


REVIEW ARTICLE OPEN



The efficacious of AOP-based processes in concert with electrocoagulation in abatement of CECs from water/wastewater

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Combining electrocoagulation with another process is a potential strategy for increasing the efficiency of water and wastewater pollutant removal. The integration of advanced oxidation processes (AOPs) and electrocoagulation (EC) demonstrates improved performance. The mechanism of the EC combined with ozone (O₃), hydrogen peroxide (H₂O₂), sulfate radicals, electrooxidation (EO), Fenton/electro-Fenton, and UV is discussed. This review sheds light on EC-AOP hybrid processes in terms of their mechanisms, development, challenges, and their potential application for the removal of contaminants of emerging concern (CECs). The majority of the articles claimed improved performance of the EC process when combined with AOP as a pre-treatment, especially in terms of removing recalcitrant contaminants. For instance, the integrated EC-Fenton/photo-Fenton processes have been shown to be a promising treatment to virtually complete removal of the phenolic compounds in oil refinery wastewater. In EC-EO process, boron doped diamond (BDD) anode, despite being costly electrode, has the highest oxidation potential and is therefore the most suitable type for the mineralization of organic pollutants. PFASs are more effective at being removed from water through zinc and Ti₄O₇ electrodes in EC-EO treatment. Furthermore, the peroxone and synergistic effects between O₃ and coagulants played almost equal dominant role to removal of ibuprofen using hybrid EC-O₃. However, enough data for conducting these integrated processes at industrial scale or with real wastewaters do not exist, and so there is a lack for comprehensive and systematic approaches to address complexity of such systems. Although a great number of papers were focused on the degradation of effluents from different industries, viruses, and pharmaceuticals, there is not sufficient research in terms of the removal of herbicides, pesticides, microplastics, and micropollutants.

npj Clean Water (2023)6:30; <https://doi.org/10.1038/s41545-023-00239-9>

INTRODUCTION

Over the past few decades, a rise in the number of harmful pollutants has been observed as a result of industrial expansion and population growth. Contaminants of emerging concern (CECs), including personal care products, medicines, pesticides, flame-retardants, plasticizers, endocrine disruptors, surfactants, and polycyclic aromatic hydrocarbons (PAHs), and food additives have become an indispensable requirement. Despite their wide-ranging uses, CECs can also be detrimental to human health and other animals. Recently, the scientific community and regulatory authorities have become increasingly concerned about CECs' widespread presence in surface and ground water sources due to their extensive use¹.

The utility of electrochemistry in drinking/waste water treatment can be a cost-effective option. The majority of contaminants in water or wastewater may be eliminated or transformed to non-toxic forms using one or a combination of electrochemical methods. Electrochemical technologies have reappeared in the last three decades, due to cheaper power costs and stronger environmental rules and norms. The electrocoagulation process is the oldest and most widespread of the electrochemical processes

used in water and wastewater treatment. Many water pollutants, such as anionic contaminants¹, phenol², sulfite and sulfate³, oil emulsions⁴⁻⁶, and anionic toxic colors^{7,8}, were successfully removed using this process. It has also been shown to be a capable method for removing heavy metal ions⁹⁻¹¹ from industrial effluents, including chromium^{12,13}, copper¹⁴, nickel¹⁵ and arsenic¹⁶, boron¹⁷, zinc, manganese and mercury¹⁸, cadmium¹⁹, lead and silver²⁰. Numerous reviews highlight the strengths of electrocoagulation (EC) technology for water and wastewater treatment²¹⁻²⁷. This method has many advantages such as a minimal setup time, no requirement for chemicals to improve removal rate, high filterability of the generated sludge, high removal efficiency, and a simple and quick control procedure. On the other hand, it has several disadvantages, including: a reliance on non-renewable resources for electrical energy, which can be mitigated with the aid of using renewable resources like solar power²⁸ and biogas²⁹, and also the use of a costly medium for oxidation (sacrificial anode) in the treatment process. According to the literature³⁰, adequate information for scaling up this technique from lab- to bench/industrial- scale is not provided.

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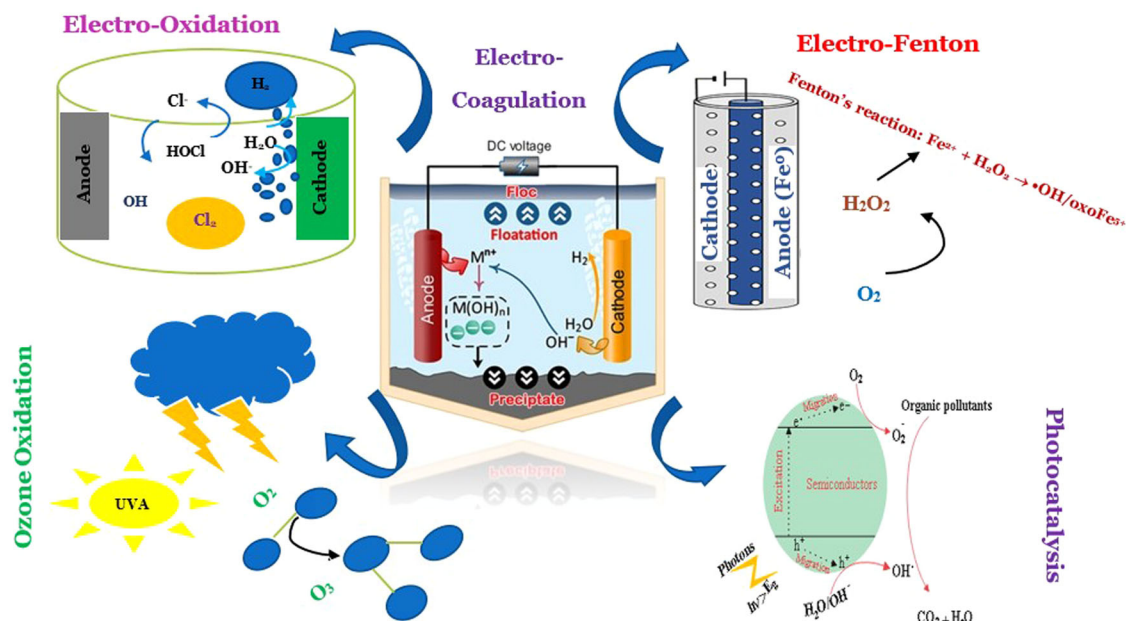


Fig. 1 Schematic representation of different AOP processes coupled with EC for CECs removal from water/wastewater. The schematic illustrates main mechanisms of electro-oxidation, Electro-Fenton, ozone oxidation, and photocatalysis integrated with electrocoagulation process for enhanced removal of CECs.

As a result, combining this process with other methods can enhance its effectiveness^{31–34}.

CECs' degradation can be enhanced by advanced oxidation processes (AOPs)³⁵. Recalcitrant pollutants in environmental matrices can be effectively treated by AOPs³⁶. Because of their ability to generate strong reactive oxygen species, advanced degradation of plentiful categories of pollutants by different AOPs has attracted tremendous attention from various researchers^{37,38}. AOPs can convert contaminants into biodegradable intermediates or result in the complete/ultimate mineralization of contaminants^{39,40}. Despite their advantages, AOPs have some drawbacks, such as forming more stable and toxic intermediates, the use of energy and chemicals, and the increase in treatment time and costs^{41,42}, which can be reduced by adding electrocoagulation to the treatment process (different AOPs coupled with EC can be seen in Fig. 1).

Nevertheless, in terms of the removal of CECs and recalcitrant pollutants electrocoagulation may lack the required efficiency potential. For instance, it is declared that penicillin-based drugs cannot be degraded completely by EC, while AOPs are very effective for this reason⁴³. It can thus be considered that, for treating highly polluted effluents and complete degradation of recalcitrant contaminants, EC integration with AOPs such as electrochemical oxidation, ozonation, Fenton, Electro-Fenton, hydrogen peroxide or sulfate radical addition, and UV irradiation can be a potential treatment option. As can be seen in Fig. 2, the terms associated with the applications of different AOPs and emerging contaminants removal are illustrated with the use of the VOS Viewer software. The data was recorded by ScienceDirect using the keywords "electrocoagulation", "advanced oxidation processes", "emerging contaminants", and "water/wastewater treatment". This issue has also attracted the attention of many researchers in the recent decade, especially since 2019 as it can be seen in Fig. 3a. What's more, the percentage of research papers published in the recent decade, based on the AOPs that was incorporated into EC, is presented in Fig. 3b.

However, there are not much available data for the performance of such processes regarding the removal of newly concerned pollutants including microplastics, viruses, and pharmaceuticals since the topic is a state-of-art. Therefore, this comprehensive review focuses on integrated EC-AOPs processes

in the last decade to shed light on the improvement in reactors design, mechanisms involved in the removal of emerging contaminants, as well as techno-economic analysis, crucial operating parameters, and kinetic models.

HYBRID ELECTROCOAGULATION-ELECTROOXIDATION

Typically, the electrocoagulation process (Fig. 4) is accompanied by flotation, in which the anodic reaction of coagulation is followed by the hydrogen production at the cathode and brings the flocs to the top of the water/wastewater. In this case, the process is called electrocoagulation/flotation. Anode and cathode materials are usually iron or aluminum and the reactions occurring around the anode are as follows⁴⁴:

For aluminum anode:



At alkaline conditions:



At acidic conditions:



For iron anode:



At alkaline conditions:



At acidic conditions:



Cathodic reaction:



Moreover, throughout these reactions there is also the possibility of oxygen evolution nearby the anode according to

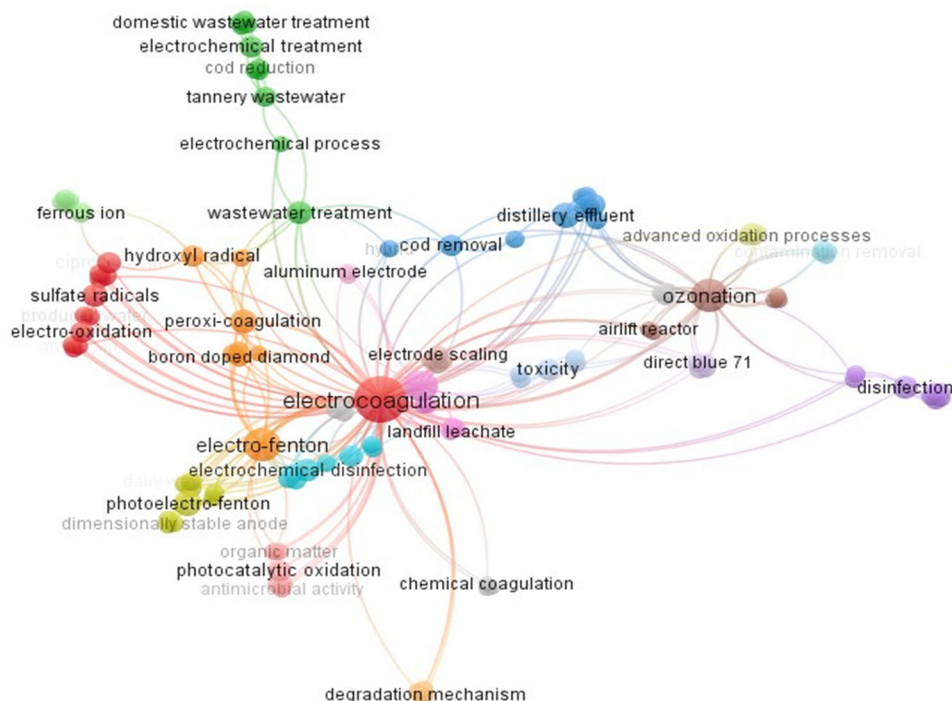


Fig. 2 Visualization network map of the keywords in the publications surveyed from the ScienceDirect published from 2012 to 2022. The network map shows the target searched keywords including integrated processes, advanced oxidation, electrocoagulation, emerging contaminants and the relationship of each with the central keyword by lines.

the following the reaction:



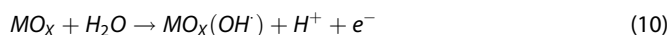
It should be noted that for the iron electrode, another mechanism may also occur during which Fe^{3+} is produced. Al^{3+} and Fe^{2+} ions are strong coagulants that absorb most of the pollutants.

Electrooxidation is the oxidation of contaminants either by the formation of hydroxyl radicals (OH^\bullet) on the surface of anode or producing oxidants in the solution⁴⁵, as shown in Eq. 9⁴⁵:



The anode material is the most important factor in electro-oxidation⁴⁶. According to the anode material, the mechanism of oxidation can be divided into indirect electro-oxidation and direct electrochemical oxidation. In the former partial oxidation occurs whereas the latter can advance complete mineralization^{29,47}. In the direct electrolysis, also named electrochemical oxygen transfer reaction, contaminants are oxidized after adsorption on the anode surface only by help of the removed electron and then an adsorbed organic radical is generated. Conversely, in the indirect electro-oxidation, also called mediated anodic oxidation, chemical oxidants are formed in-situ either by anodic oxidation such as generation of active chlorine, ozone and persulphate, or by cathodic reduction such as generation of hydrogen peroxide⁴⁸.

The first stage of both mechanisms is the release of water molecules at the anode surface to produce adsorbed hydroxyl radicals as presented in Eq. 10⁴⁵ where MO_x represents the metal oxide anode.



The subsequent steps depend strongly on the nature of the anode materials, which is classified into "active" and "non-active" anodes for indirect electrochemical oxidation and direct electro-oxidation, respectively. The active anodes, used in indirect electro-oxidation, have higher oxidation states resulting in firmer

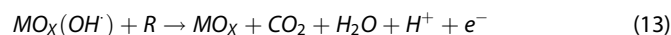
interaction with the adsorbed hydroxyl radicals by further oxidation, forming higher oxides (Eq. 11⁴⁵).



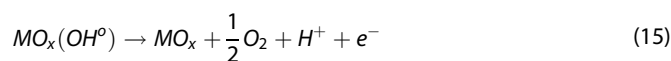
The surface redox couple MO_{x+1}/MO_x , denoted as chemisorbed active oxygen, is the mediator of the partial oxidation of organics on active anodes. As demonstrated in Eq. 12⁴⁵, it is obvious that the anode returns to its original oxidation state, thus these are called non-sacrificial anodes. Generally, anodes with low over-potential of oxygen growth such as amorphous carbon, graphite, platinum, ruthenium dioxide, and iridium dioxide are categorized as "active" electrodes.



On the contrary, the non-active anodes are defined by a weak electrode hydroxyl radical interaction creating physisorbed active oxygen, which promotes the complete mineralization of the organics to carbon dioxide (Eq. 13⁴⁵).



Though, a competitive oxygen evolution side reaction (Eqs. 14–15⁴⁵) happens in both of the chemisorbed and physisorbed active oxygen evolution mechanisms and results in reduced anodic process efficiency and waste of energy. Mainly, electrodes with high over potential of oxygen evolution such as lead dioxide, antimony-doped tin oxide, or boron-doped diamond (BDD) are entitled as "non-active" anodes.



The whole reactions of both mechanisms of electro-oxidation are represented in Fig. 5 schematically as a full oxidation cycle⁴⁹.

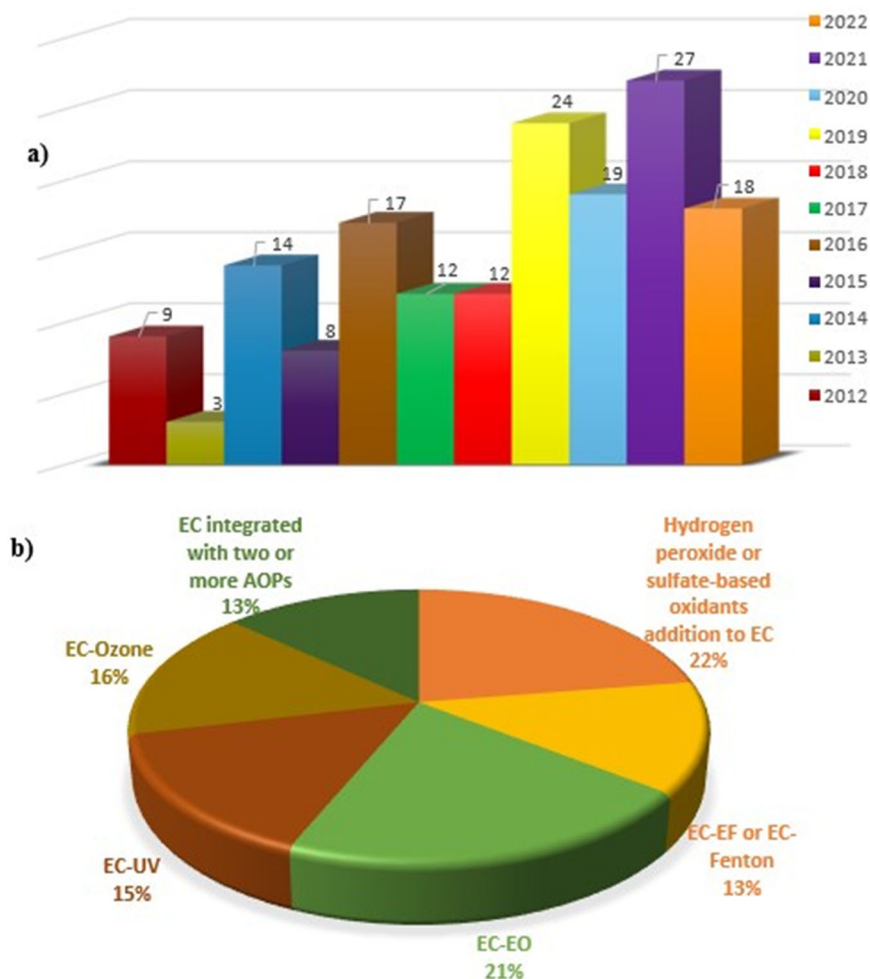


Fig. 3 Bibliometric study regarding integrated EC and AOPs on CECs removal between 2012 and 2022. **a** Number of original articles published annually on the application EC-AOP. **b** Percentage of each AOP-based processes integrated with EC according to ScienceDirect (assessed: 27 August 2022).

The weak involvement of hydroxyl radical with non-active anodes imposes a need for great anodic potential for water oxidation (approximately 2.0 V per SHE)⁵⁰. On the other hand, oxidation states cycle repeatedly in the electrochemical oxidation of the active electrodes. The BDD anode has the highest oxidation potential and is therefore the most suitable type for the mineralization of organic pollutants. The higher the over potential, the weaker the hydroxyl radicals in the anode will be and less energy will be used for the side reaction of water oxidation. The activity of "Oxygen evolution reactions" for Pt, IrO₂-Ta₂O₅ and RuO₂-TiO₂ are higher which makes them less effective in pollutant mineralizing⁵¹.

As noted above anode material is the most important parameter in the process. In summary, advantages and disadvantages of each anode type is as follows⁵⁰:

- Graphite is cheap but ineffective and unstable.
- Platinum is expensive with low-impact.
- IrO₂ and RuO₂ are only effective for the mediated anodic oxidation.
- Due to the low conductivity, SnO₂ must be doped with Sb which is a toxic substance.
- PbO₂ is available and easily made, but unstable and should be doped with Bi, Co or Fe or mingled with Teflon.
- TiO₂ has low conductivity and often Ebonex® is used in electrooxidation. Ebonex® is a non-stoichiometric titanium oxide mixture contained of Magneli phase titanium oxides

Ti₄O₇ and Ti₅O₉. It has a rather difficult and expensive way to be made and surface passivation under extreme settings may occur.

- BDD is stable and effective but very expensive.

Another drawback of electrooxidation is that it works poorly when purifying effluents with a high concentration of suspended solids. In this case, suspended solids must first be treated using other methods, which can be accomplished by combining it with other techniques. Coupling electrocoagulation with electrooxidation provides a feasible combination that takes advantage of both processes. When colloidal particles are present, electrooxidation becomes impractical due to its significant operating time consumption. Electrocoagulation is used to speed up the process and remove colloidal elements and charged types from the water. While electrocoagulation may remove pollutants rapidly, it cannot do so entirely, whereas electrooxidation is a slow process, but it removes contaminants consistently⁵². Combining the benefits of two processes that provides quick and total pollutant removal is possible through sequential and simultaneous hybrid systems.

Electrocoagulation and electrooxidation are the most promising electrochemical treatment methods due to their low or no chemical needs and ease of use⁵³. To study the effectiveness of the hybrid EC-EO process for the treatment of each type of water/wastewater, usually a batch mode is used. Once a batch system is employed to find the ideal operating parameters, a continuous

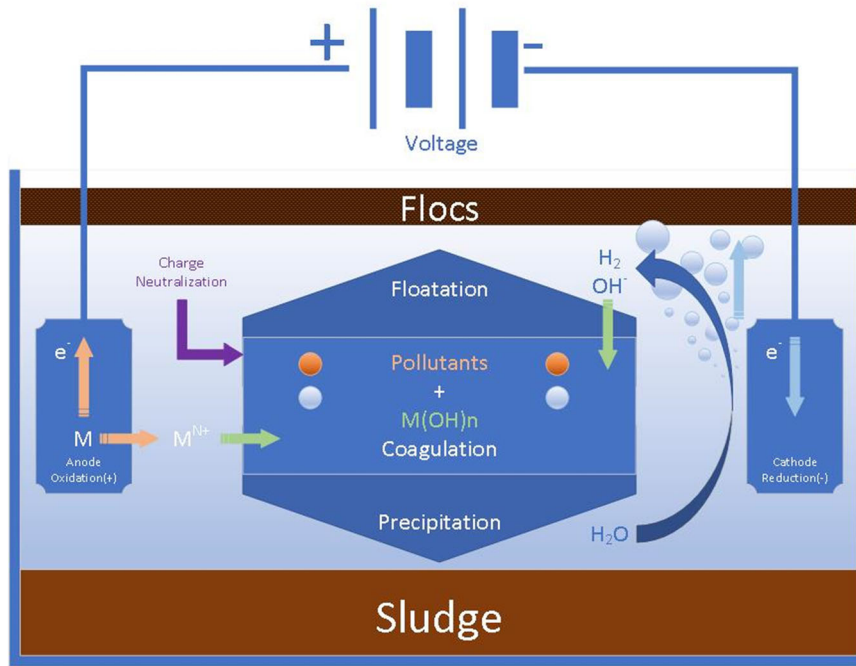


Fig. 4 Main mechanism involved in electrocoagulation process. A current is passed through a metal electrode. This process involves the dissolution of metal cations from the reactor anode with the simultaneous formation of hydroxyl ions and hydrogen gas at the cathode.

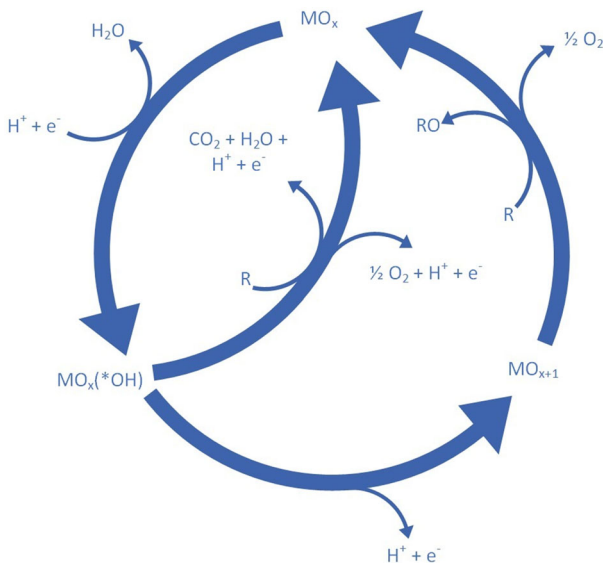


Fig. 5 General scheme describing the reaction mechanisms of electrooxidation of organic compound. Such mechanisms including formation of hydroxyl radicals (OH^*), oxygen evolution by electrochemical oxidation of OH^* , formation of the higher metal oxide (MO), oxygen evolution by chemical decomposition of the higher MO, electrochemical combustion of the organic compound via OH^* , and electrochemical conversion of the organic compound (R) via the higher MO. Reproduced with permission from ref. ⁴⁹. Copyright 2006 Royal Society of Chemistry.

mode can be used to find out the effects of pollutant concentration and flow rate and to see whether there is a correlation⁵⁴. A few studies have compared batch and continuous reactors for integrated form of electrocoagulation and

electrooxidation⁵⁵. For instance, Lakshmi Kruthika et al.⁵⁶ used a hybrid method with continuous and batch reactor systems. Using continuous and batch reactor systems, the highest removal of total organic carbon (TOC) was attained in the range of 38–54% and 80%, respectively. Linares Hernandez et al.⁵⁷, on the other hand, investigated soft drink wastewater treatment in a batch-type reactor using an integrated of successive electrocoagulation and electrooxidation processes. As a result, at pH 8, only 27% and 85% of TOC could be eliminated by EC and EO, respectively.

For the treatment of a textile wastewater, Afaha et al.⁵⁸ disclosed that the hybrid treatment method of EC-EO is a promising method since the COD reduction percentage rose from 76% to 97%. In a study for treating soluble coffee industrial plant wastewater⁵⁹, an amazing formulation was also derived for estimating the total cost of the sequential process of EC-EO. In addition, anode material consumption (AMC), energy consumption, and sludge formation have all been taken into account not only for electrocoagulation but for electrooxidation. A linear increase in AMC was observed with the extension of the EC operation time (t_{EC}). Anode material consumption increases linearly with applied current, according to Faraday's Law (Eq. 16):

$$AMC(\text{kg}/\text{m}^3) = 0.0138 \times t_{EC} \quad (16)$$

Another example of treatment by sequential EC-EO is the treatment of container washing wastewater by Nayr and Kara⁵³. They observed that EC performed less well in the removal of soluble chemical oxygen demand (sCOD) than expected and so EO was used as a post-treatment to EC. As removal efficiency of color declined at the same length of time in the hybrid process, they inferred that EO was not practical and related it with aqueous solution's colloidal particles. However, a very recent study in Canada on a similar wastewater, reached different results⁶⁰. Azarian et al.⁶¹ observed that solo EC process was unable to treat the tannery effluent completely, but when electrooxidation was used as the post-treatment, results were very satisfactory. The question of whether EC should be utilized as the pre-treatment or post-treatment to EO was examined for

Table 1. Applying integrated EC-EO process for water/wastewater treatment.

| Ref. | Electrode materials | Removal rate | Comments | Wastewater/Pollutant |
|------|---|--|--|--|
| 191 | EC: Al EO: Ti/RuO ₂ | 34.5% COD 83.9% NH ₃ -N | Current density = 40 mA/cm ² EC time = 30 min EO time = 360 min | Landfill leachate |
| 56 | EC: Al EO: TiO ₂ nanotube | EC: 60% TOC EO: 80% TOC | Enhanced oxidation was detected with TiO ₂ nanotube electrode | Gelatin production plant |
| 65 | Simultaneous hybrid process-SS (MP) | 84% Diclofenac Sodium 80% COD | pH = 10.9 | Synthetic pharmaceutical wastewater |
| 66 | EC: Fe EO: BDD | 97% phosphorus 95% COD | Current density = 0.08 A/cm ² Electrolysis time = 90 min | Industrial container wash water |
| 52 | EC: Al EO: SS | 93% COD 93% TOC 98% Phenol 94% O&G | Electrolysis time = 20 min Current density = 16.6 mA/cm ² | Petroleum refinery |
| 61 | EC: Al EO: Pb/PbO ₂ | 98.9% COD (≤ 5 mg/L) 99% Color | Energy consumption in hybrid process = 18.2 kWh/m ³ Energy consumption in EC = 60 kWh/m ³ EC time = 10 min EO time = 40 min EC Current density = 17 mA/cm ² EO Current density = 42 mA/cm ² | Tannery |
| 192 | EC: Al/Fe (BP) EO:MP graphite | 87% COD 100% Color and Turbidity | Energy consumption = 31.6 kWh/kg COD Current intensity = 1A pH = 6 | Real pulp and paper mill |
| 53 | EC: Fe/Al EO: BDD | 89% CODs 72% Color | Color removal efficiency declined in the hybrid process EC time = 120 min EO time = 420 min | Container washing |
| 193 | EC: Fe EO: BDD | 97% COD Complete virus inactivation | The first attempt for virus mitigation by sequential EC-EO | Drinking water |
| 194 | EC: Fe EO: PbO ₂ | 95.6% COD 90.5% TOC 91.6% BOD 99.8% NH ₄ -N | EC + EO + peroxymonosulfate (PMS)/UV/CuFe ₂ O ₄ for treating highly toxic landfill leachate | Landfill leachate |
| 67 | EC: Fe EO: BDD | 56% E3 64% for EE2 | EC current density = 8.88 mA/cm ² EO current density = 22.2 mA/cm ² EC and EO time = 8 and 40 min pH = 6 | Estrogenic compounds in municipal wastewater |
| 195 | EC: Fe/Al/SS EO: BDD | 90.1% COD 88.7% Phenol | Penicillium glabrum had the highest COD and total phenol removal efficiency compared to other fungal strains | Pistachio processing |
| 64 | EC: Zn/SS EO: Ti ₄ O ₇ | 99% perfluorooctanesulfonic acid 99% perfluorononanoic acid 99% fluorotelomer sulfonic acid 99% PFOA 90% perfluorohexanesulfonic acid 90% fluorotelomer sulfonic acid | First attempt for treating PFASs by EC-EO Investigated foam generation and floc morphology | Per- and polyfluoroalkyl substances (PFASs) |
| h | EC: Al EO: Al/cathode and Ti/IrO ₂ /anode | 100% NH ₄ ⁺ -N 96.3% Nitrate | Al/cathode and Ti/IrO ₂ /anode electrodes were applied to oxidize NH ₄ ⁺ to N ₂ . pH = 7.5 Current density = 30 mA/cm ² | Natural groundwater |
| 196 | EC: Fe EO: Ti/SbO ₂ | 95.4% COD 89.4% NH ₃ -N 99.99% E. coli | EC time = 20 min EO time = 30 min EC Current density = 100 A/m ² EO Current density = 100 A/m ² | Municipal wastewater |

treating poultry slaughterhouse/dairy wastewaters⁶². Descriptions of reviewed paper in this scope are provided in Table 1.

Landfill leachates as one of the most concerning problems of the current era are proved to be very toxic and difficult to degrade. In landfill leachate treatment, electrochemical techniques have gotten a lot of interest during the last five years⁶³. In a recent work, to remove and degrade polyfluoroalkyl substances (PFASs) from water, Shi et al.⁶⁴ combined electrocoagulation with electrooxidation, using zinc and stainless-steel in the

former and Ti₄O₇ electrode in the latter. In Fig. 5, concentration profiles of three different PFAS-containing solutions that went through EC under various conditions are shown. PFASs elimination through EO is obvious for all of the EC-derived solutions. More than 95% PFAS removal was attained in 60 min of EO for Solution I (Fig. 6a), excluding PFBS (73.4%), and it was over 90% for Solution III (Fig. 6c) bar PFBS (52.3%), PFHxA (74.2%) and 4:2 FtS (85.9%). Degradation of all kinds of PFAS in Solution II (Fig. 6b) was satisfactorily high. Overall, they found that long-chain

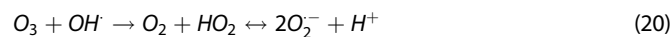
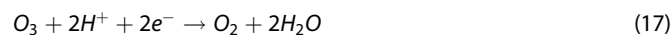
PFASs (C7–C10) was more effective at being removed from water by electrocoagulation with zinc anodes. As a consequence of the high current density and/or the high PFAS concentration during EC, foam was produced, promoting the separation of PFASs from the bulk solution, specifically for the long-chain PFASs. Also, viruses and bacteria in water and wastewater can be inactivated by sequential EC-EO^{65,66}.

Maher et al.⁶⁷ investigated four resistant estrogenic compounds, namely estrone (E1), 17 β -estradiol (E2), estriol (E3), and 17 α -ethynylestradiol (EE2) and found that for each estrogenic compound, the sequential EC and EO method was efficient by more than half over EO alone. The results revealed that EC as the pretreatment also reduced the energy consumption. Pesticide-containing effluent is another concern that has been shown to be treated very well by electrochemical processes^{68–71}.

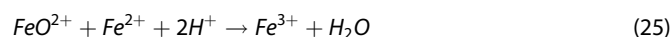
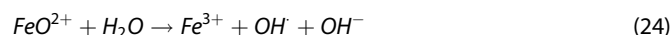
HYBRID ELECTROCOAGULATION-OZONE

The oxidation of numerous organic and inorganic compounds by ozone can be performed via two main mechanisms: direct oxidation as well as indirect oxidation using the free radicals' generation

including the hydroxyl radical (OH \cdot), as shown by the Eqs. 17–22⁷²:



Electrocoagulation (EC) coupled with ozone (O₃) bubbles involves redox processes in which ozone decomposes into FeO²⁺ and hydroxyl radical (OH \cdot) via reactions with Fe²⁺ ions and Fe³⁺ ions⁷³. The following are the redox reactions that take place:



By adding radicals to organic compounds, hydrogen abstraction, and transferring electrons to the hydroxyl radicals, as a result

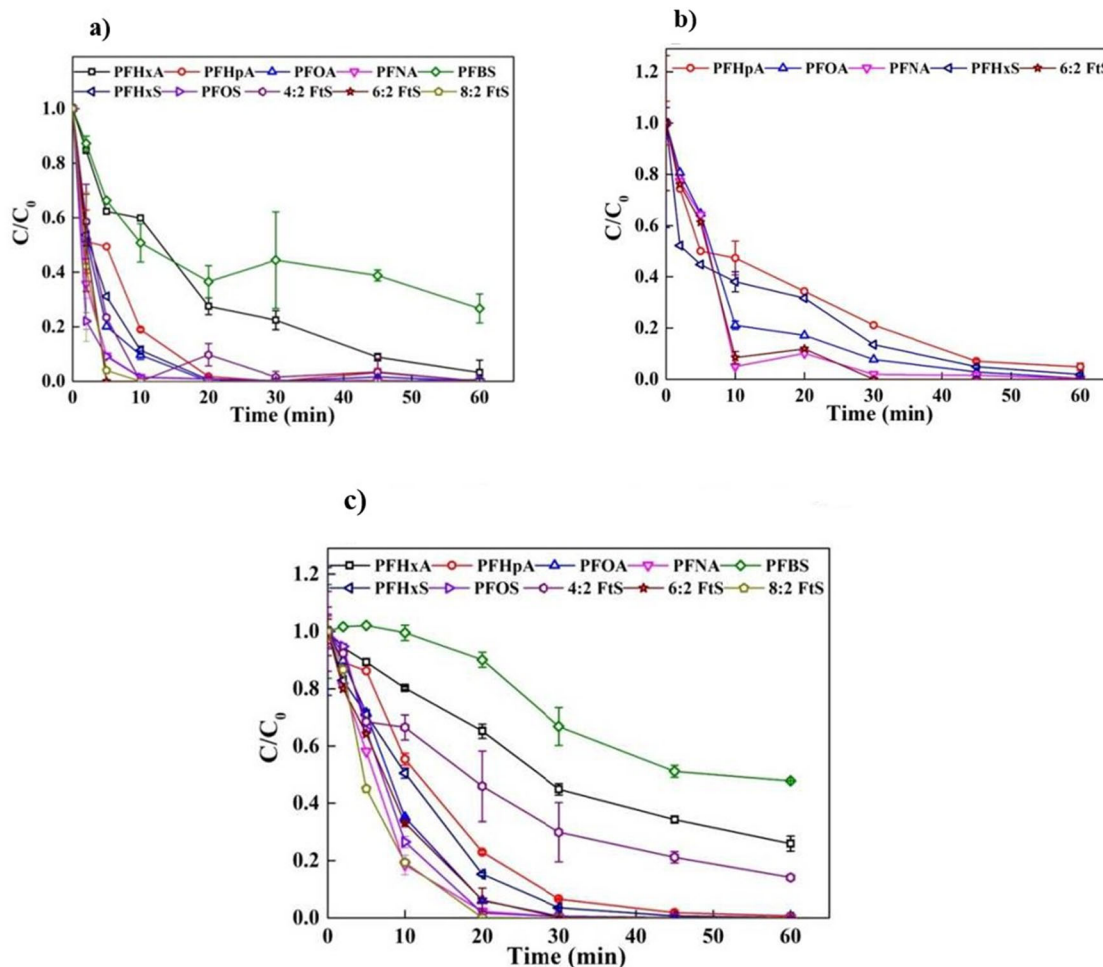


Fig. 6 Concentration profiles of three solutions after EC and during EO with current density of 10 mA.cm⁻². **a** Solution I: PFASs-laden flocs formed by EC at 0.3 mA.cm⁻² with dissolved acid; **b** Solution II: PFASs-laden flocs prepared by EC at 5 mA.cm⁻² with dissolved acid; **c** Solution III: foam made throughout EC at 5 mA.cm⁻². Reprinted with permission from ref. ⁶⁴. Copyright © 2021 Elsevier Ltd.

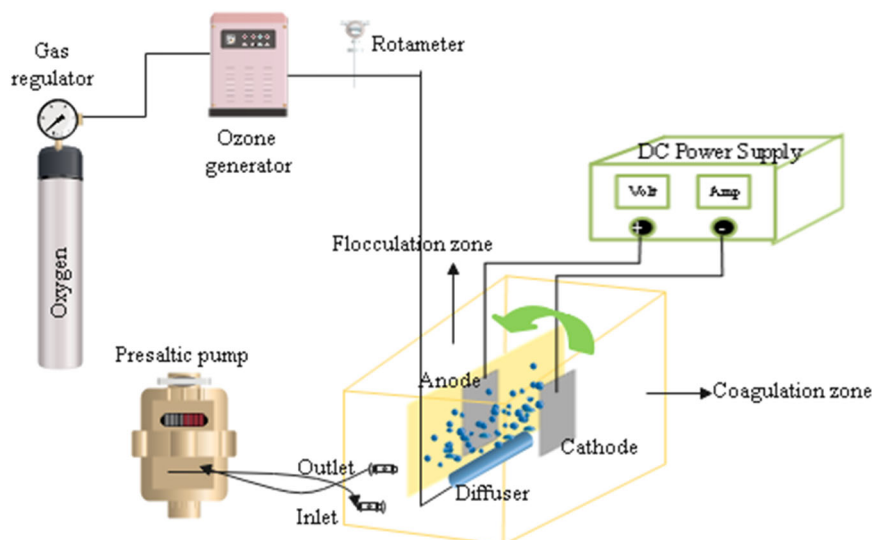


Fig. 7 Schematic experimental EC-O₃ setup with a novel reactor design. The EC-ozonation reactor with a middle wall was used to reduce floc breakage under ozonation-induced excessive turbulence. Reproduced with permission from ref. ⁸⁰. Copyright © 2021 Elsevier Ltd.

the organic compounds of the medium are oxidized due to the hydroxyl radicals⁷⁴. Equations 23–25 demonstrate a mutual activation between Fe²⁺ and O₃. In EC mediums containing Fe²⁺, ozone decomposes more quickly to hydroxyl radicals. O₃ bubbles within an EC system produce a catalytic O₃/Fe²⁺ that forms FeO²⁺ which is an intermediate species reacting with water to produce hydroxyl radicals, HO ions, and Fe³⁺ covalent ions. Organic pollutants can decompose more rapidly through these mutual activation products, thus improving process efficiency⁷⁵. The combination of O₃ and EC treatment processes, especially for the removal of organic pollutants, has therefore been the subject of a wide range of research, the first of which was that of He et al.⁷⁶ who tested the elimination of C.I. Reactive Yellow 84 by adding ozone to electrocoagulation process.

In order to treat real distillery industrial effluents, Asaithambi et al.⁷⁷ used three distinct methods: EC, O₃ and integrated EC-O₃. In the hybrid process of EC-O₃, in-situ active radicals attack and oxidize the organic substance as a result of the interaction between ozone and Fe²⁺ ions created by iron electrode oxidation, as indicated by Eqs. 6–8. In addition to chemical oxygen demand (COD), color was significantly lowered as a result. They reported similar observations for the COD removal from actual distillery wastewater⁷⁸ and landfill leachate⁷⁹. It is of concern that EC or the combinations of O₃-EC processes outperformed ozonation standalone in removing both color and COD. The inadequate mass transfer of ozone from the gaseous phase to the liquid in which the organic contaminants might be the cause.

Ahangarnokolaei et al.⁸⁰ proposed EC-O₃ reactor (Fig. 7), which is enhanced coagulation under O₃-induced rapid mixing in the first side, as well as flocculation under flow-induced slow mixing on the other side. It lowered floc degradation at high O₃ dosages and improved removal rate. For integrated processes, especially simultaneous combination, superior performance was observed with less treatment times. When utilizing Al electrodes, the synergistic impact of EC and O₃ on dye, COD and TOC removal was found to be more pronounced because of the increased ozone activation and decreased anode corrosion. They suggested the simultaneous process of EC (Al)-O₃ and sequential process of EC (Fe)→O₃ in their suggested reactor design as an innovative and appropriate solution for treating dye wastewater.

The O₃-assisted EC process was investigated by Bilinska et al.⁸¹ for the removal of Reactive Black 5. The high pH value of the water in the EC process lowered the efficacy of the aluminum electrodes, making the iron electrodes better conductors. In addition, a semi-

batch stirred cell that held 1 L of 1 g/L ozone was tested and the color removal rate was 90% after 50 min. The O₃ time was reduced from 50 to 10 min, saving five times as much as it would have saved had it not been pretreated with EC. Similarly, García-Morales et al.⁸² conducted a study for the dye removal in Denim wastewater and 66% color removal was reported when EC-O₃ was run. According to their findings, second-order kinetics for color and turbidity removal in the O₃-EC pulses method indicated that the efficiency achieved in the integrated process was dependent on the coupling of treatments (O₃ and EC). Tanveer et al.⁸³ also investigated the efficacy and practicality of EC/O₃, EC/Fenton, and EC/photo-Fenton processes for the treatment of textile wastewater and demonstrated effective decolorization and removal of TDS, TSS, and COD from textile dye-bath effluents compared to the solo EC process. EC alone was able to eliminate 57% COD, but when combined with ozone the removal percentage reached to 77%. Aseman-Bashiz and Sayyaf⁸⁴ concluded that solely EC could not profoundly effect on ofloxacin removal (only 19% after 90 min). Therefore, integrated process through O₃ and peroxydisulfate was achieved higher removal rate of 98% under optimum conditions. Most importantly, such process easily coped with conventional issues of EC such as sludge production and electrode corrosion.

To investigate the effectiveness of EC/O₃ in treating greywater, Barzegar et al.⁸⁵ used a hybrid approach and observed that when the pH of greywater was near to neutral, the system worked effectively. In contrast to EC with Al electrode, EC with Fe electrode displayed strong catalytic activity for O₃ activation. Also, when used in combination with the EC method for greywater treatment, O₃ performed better than other chemical oxidants (peroxydisulfate, peroxymonosulfate, and hydrogen peroxide). Jin et al. conducted a research⁸⁶ on the removal of ibuprofen by EC-O₃ hybrid system showed that the peroxone (O₃/H₂O₂) and synergistic effects between O₃ and coagulants (SOC) had almost equal contribution to ibuprofen removal (Fig. 8). Also of note, the study of Das et al.⁸⁷ that concentrated on the removal of COD, BOD, cyanide, and chloride from biological oxidation treated effluent of the steel plant. Results revealed that a rise in the volumetric mass transfer coefficient, K_{1a}, as ozone production rate rose.

It is also of notice that the electro-peroxone (E-peroxone) technique is an innovative update of the ozonation that generates hydrogen peroxide in-situ during ozonation from cathodic oxygen reduction at carbon-based cathodes (Eq. 26)⁸⁸. The in-situ

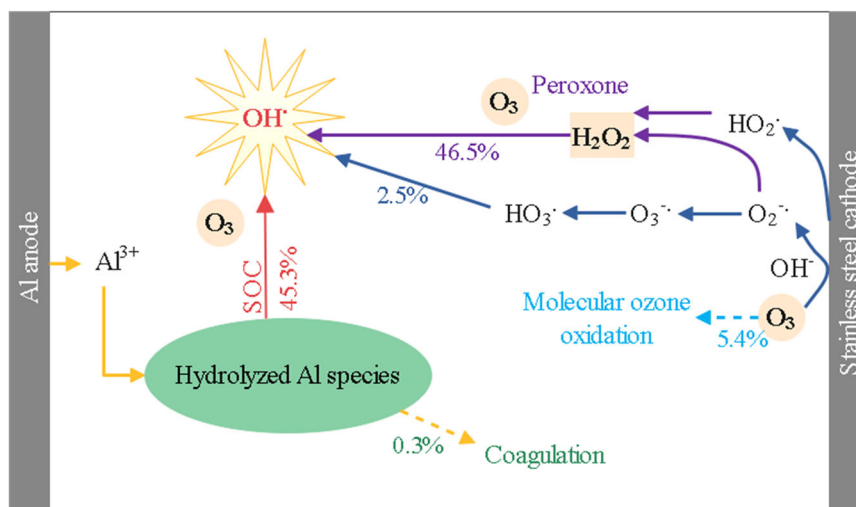
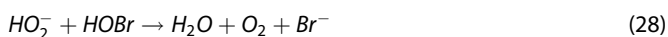
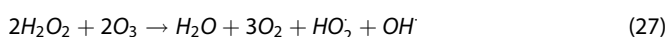


Fig. 8 Synergistic effects between ozone and coagulants (SOC) are involved in the E-HOC. Based on the reactive oxygen species (ROS) detection and reactions on the electrodes, the synergistic effects between ozone and coagulants (SOC) were found to be involved in the E-HOC process. Reproduced with permission from ref. ⁸⁶. Copyright © 2020 Elsevier Ltd.

produced H_2O_2 can then be used through the peroxone reaction in order to boost the transformation of O_3 into OH^\cdot (Eq. 27)⁸⁸. Furthermore, compared to conventional O_3 , the E-peroxone process can dramatically reduce bromate formation in the treatment of bromide-containing water because O_3 possesses shortened lifespan and quenching of hypobromous acid, which is a key intermediate for bromate formation in the O_3 -induced process, by electro-generated H_2O_2 (Eq. 28)⁸⁹. When compared to ozonation, the E-peroxone technique can considerably enhance efficiency and decrease energy consumption for the removal of O_3 -resistant pollutants owing to the improvement of O_3 transition to hydroxyl radicals by electro-generated H_2O_2 . As a result, the E-peroxone method can significantly strengthen the performance of water and wastewater treatment by simply putting electrodes in O_3 reactors⁹⁰. Wang et al. treated shale gas fracturing flowback water by the sequential EC-Eperoxone (Fig. 9) and concluded that adding E-Peroxone after the EC process increased both current efficiency and enhancement factor (Eq. 55)⁸⁸. The latter implies that hydrolyzed Al species could have a synergistic effect in the ECP process, accelerating O_3 decomposition to create hydroxyl radical. Hydrogen peroxide can also be added to the system instead of being generated in-situ⁹¹. All papers reviewed in this area are summarized in Table 2.

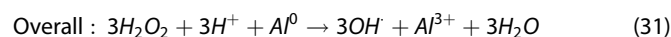
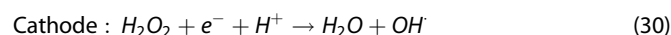


Many obstacles remain, however, preventing large-scale implementation of ozone-assisted electrocoagulation since there are far more significant issues regarding mass transfer of ozone gas to the liquid phase. As a result, finding a proper reactor design that facilitates high rates of ozone mass transfer to the liquid phase is the next step. The novel reactor system was employed by Ahangarkolei et al.⁸⁰ (Fig. 7) are good attempts at achieving an appropriate reactor design, as demonstrated by the results. However, in a circular cross section reactor, mixing among the phases will be more effective than in a square cross section, which might lead to findings that are more positive. Secondly, the lack of any scale-up attempts for ozone-assisted electrocoagulation process is a major problem because the reactor design is crucial in optimizing the overall performance of the process and it often

incorporates several phases, including the gas phase. Ozone generation on a large scale is also another problem that might be overcome by the use of electrochemical technology to create oxidants and simultaneous anodic oxidation⁹².

PEROXI-COAGULATION, AND HYBRID ELECTROCOAGULATION-FENTON/ELECTRO-FENTON PROCESSES

In the Peroxi-electrocoagulation process, pollutants are eliminated by generating hydroxyl radicals owing to adding of H_2O_2 during the conventional electrocoagulation (EC). In this technique, organic pollutants are converted to H_2O , CO_2 and inorganic ions⁹³. When hydrogen peroxide is introduced to the anode and cathode surfaces during electrocoagulation, in which Al electrodes are used, the following reactions occur⁹⁴:



This causes chain reaction between the hydroxyl radical and an organic matter (R) and so oxidizes organic pollutants according to the following reactions⁹⁵:



When iron is used instead of aluminum, the mechanism is different. By incorporating the Fenton reagent into an oxidation process, which results in the reaction of hydrogen peroxide, H_2O_2 , and ferrous salt, Fe^{2+} , to oxidize organic matter⁹⁶. An enhanced form of the Fenton process, electro-Fenton is a method in which H_2O_2 is produced electrochemically in-situ under acidic circumstances. Cathodic Fenton processes and anodic Fenton processes are two different ways of describing the process, depending on how iron enters the system. One or both reagents $Fe(II)$ and H_2O_2 are created in-situ in cathodic Fenton reactions, when iron is supplied as $Fe(III)$ salt. The source of the ferrous ion involved in Fenton reaction in anodic Fenton processes (peroxi-coagulation) is a sacrificial iron anode^{97,98}. The followings are the primary

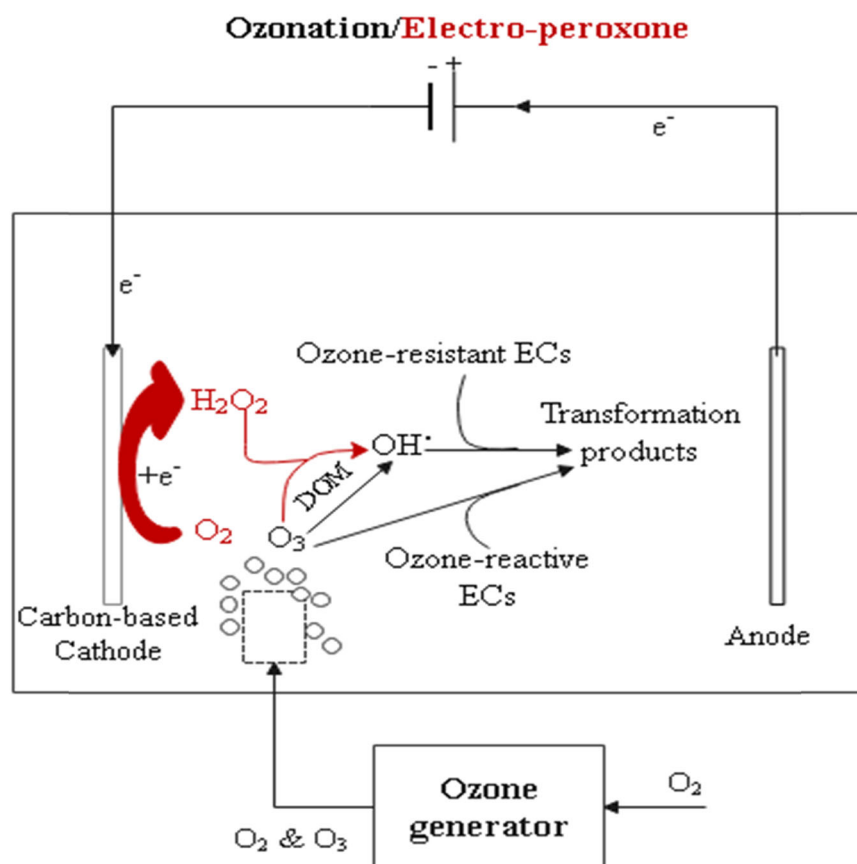
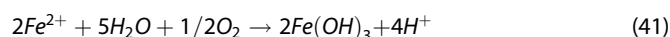
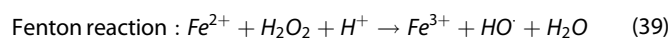
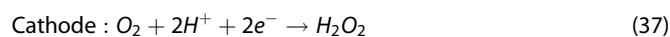


Fig. 9 Schematic of E-Peroxone process mechanism. The transformation of enhanced ozone (O_3) to hydroxyl radicals (OH^\cdot) by electro-generated H_2O_2 , the E-peroxone process can considerably increase the efficiency and decrease the energy demand for the abatement of ozone-resistant ECs. Reproduced with permission from ref. ⁸⁸. Copyright © 2018 Elsevier Ltd.

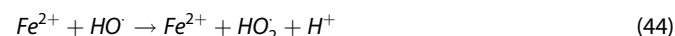
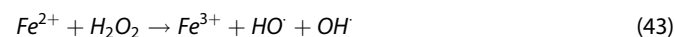
reactions that occur throughout the electro-Fenton process⁹⁹:



Fe^{2+} and H_2O_2 are generated between the anode and cathode (Eqs. 35–38) and then hydroxyl radical can be electrogenerated in the bulk solution through the Fenton reaction (Eq. 39). Due to powerful attacks on aromatic rings or long chains, the hydroxyl radical can non-selectively transform difficult-to-degrade organic contaminants into small micro molecules. Furthermore, this process allows for coagulation¹⁰⁰, which is precipitated by iron hydroxides produced as $Fe(OH)_n$ ($n = 2, 3$) (Eqs. 40–42). As a result of the combined actions of oxidation and coagulation, persistent contaminants may be more easily converted into biodegradable chemicals and even eliminated. A number of studies have shown that electro-Fenton process is more effective in acidic conditions, particularly at pH levels of 3 or below¹⁰¹. However, if pH is less

than three, H_2O_2 availability may be reduced. This leads to coagulate contaminants by $Fe(OH)_n$ produced at higher pH because of the enhanced decomposition of H_2O_2 into O_2 and H_2O due to an increase in pH¹⁰².

When hydrogen peroxide is not produced in situ and is added to the EC system, following reactions take place:



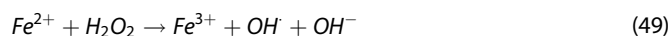
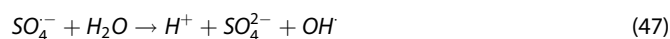
Reduction of the ferric species formed by the ferrous ion regeneration with hydrogen peroxide is the primary means of propagating this process. More ferrous ions are consumed than they are created in a certain period. Hydroxyl radicals, on the other hand, have the ability to swiftly degrade ferrous ions. In order to keep hydroxyl radical generation at a manageable level, greater ferrous ion dose is required. The neutralization stage of the Fenton process generates a substantial volume of ferric hydroxide sludge that must be separated and disposed later on¹⁰³. Persulfate has also been considered as an efficient oxidant because its redox potential is 2.6 V¹⁰⁴. Additionally, it has a longer half-life compared with hydroxyl radical due to its preference for electron transfer reaction, whereas OH^\cdot is capable of taking part in a variety of reaction with equal preference. As an example, 2,4,6-trinitrotoluene was degraded to 88% by the EC-persulfate method¹⁰⁵. Thus, it is less selective than $SO_4^{\cdot-}$ regarding the oxidation of organic pollutants¹⁰⁶. In the integrated EC/Persulfate system, Fe^{2+} is produced from the sacrificial iron electrode and also hydrogen

Table 2. Hybrid EC-O₃ process for water and wastewater treatment.

| Ref. | Electrode materials in EC | Removal rate | Comments | Wastewater/Pollutant |
|------|--|---|--|--|
| 76 | Fe | 85% TOC 97% Color | pH = 6.5 O ₃ flow rate = 20 mL/min Current density = 15 mA/cm ² | C.I. Reactive Yellow 84 |
| 197 | Fe | 50% TOC 80% Color | pH = 7 O ₃ flow rate = 90 mL/min Current density = 88.6 mA/cm ² | Acid Orange 6 azo Dye |
| 198 | Fe | 80% TOC 96% Color | pH = 10 O ₃ flow rate = 20 mL/min Current density = 10 mA/cm ² | Reactive Blue 19 |
| 199 | Fe | 100% Color | pH = 6.1 O ₃ flow rate = 500 mL/min Current density = 1.5 mA/cm ² | Red MX-5B |
| 84 | Electro-activation (Fe) of ozone and peroxydisulfate | 98% | pH = 4 Ofloxacin = 5 mg/L Current intensity = 45 mA Nanopyrite particles = 1 g/L Time = 90 min | Ofloxacin |
| 78 | Fe/Al | 100% Color 83% COD | pH = 6 O ₃ flow rate = 33.3 mL/min Current density = 10 mA/cm ² | Distillery effluent |
| 200 | Al | 79% COD | pH = 7 Electrolysis time = 12.5 min O ₃ flow rate = 60 mL/min Current density = 66 mA/cm ² Simultaneous hybrid process | Industrial park effluent |
| 82 | Al | 65% Color 76% Turbidity 37% COD | pH = 7.5 Electrolysis time = 120 min O ₃ = 5.2 g/hr Current density = 0.2915 mA/cm ² | Denim dyeing effluent (indigo carmine dye) |
| 201 | Fe/Al | 100% Fe 30% COD | pH = 7 O ₃ flow rate = 5 mL/min Current density = 25 mA/cm ² | Ripe table olive processing effluent |
| 202 | Al | 99.99% Color 99.35% COD | pH = 8.7 O ₃ flow rate = 5 g/hr Current density = 6.66 mA/cm ² | Offset printing dyes |
| 203 | Fe/Al/Cu | 88% COD | pH = 7.3 Electrolysis time = 12 min Current density = 150 A/m ² | Real industrial wastewater |
| 204 | Fe | 100% Color | pH = 9.5 O ₃ flow rate = 5 mL/min Current density = 6.66 mA/cm ² Novel design of a rectangular internal-loop airlift reactor | Acid Brown 214 |
| 137 | Fe | 97% BPA | pH=5 O ₃ = 162 mg/L Current density = 0.6 mA/cm ² | Bisphenol A |
| 77 | Fe | 100% Color 97.5% COD | pH = 7 O ₃ flow rate = 15 L/min Current density = 3.85 mA/cm ² | Distillery effluent |
| 205 | Al-graphite | 84.6% TOC 91.3% COD 92.6% BOD 90.4% Oil & grease | pH = 8 O ₃ flow rate = 9.2 g/hr Current density = 9 mA/cm ² | Greywater |
| 81 | Fe/Al | 98% Color | pH = 12 Electrolysis time = 18 min O ₃ = 0.3 g/L Ozone dose reduced 5 times when coupled with EC- EC → O ₃ was more cost-effective | Reactive Black5 |
| 79 | Fe | 97.5% COD 100% Color | pH = 7 Electrolysis time = 5 h O ₃ flow rate = 3.75 g/hr Current density = 5 A/dm ² EC(Fe)-O ₃ -Sonication process Energy consumption = 8 kWh/m ³ | Landfill leachate |

| Ref. | Electrode materials in EC | Removal rate | Comments | Wastewater/Pollutant |
|---------|---------------------------|---|--|-----------------------------------|
| 86 | Anode: Al Cathode: SS | 58.6% DOC (dissolved organic carbon) more than 70% ibuprofen | pH = 5 Electrolysis time = 10 min Current density = 15 mA/cm ² O ₃ = 10.9 mg/L | Municipal wastewater |
| 206,207 | Al | 92% Color 87.2% COD | Initial pH = 3.1 Electrolysis time = 180 min O ₃ flow rate = 3 g/hr Current density = 9.75 A/cm ² Punched Al–Al electrodes were the best | Distillery spent wash |
| 80 | Al Fe | 99.51% COD 62% TOC 99.44% COD 51% TOC | pH = 8 Electrolysis time = 30 min Electrolysis time = 50 min O ₃ flow rate = 1.06 g/hr Current density = 3.54 mA/cm ² EC(Al)–O ₃ was the best option | Direct Blue 71 |
| 87 | Al | 99.8% Cyanide 94.7% COD 95% BOD 46.5% Chloride ions | Electrolysis time = 30 min Ozonation time = 40 min O ₃ flow rate = 1.33 g/s Current density = 100 A/m ² | Real steel industry wastewater |
| 208 | Fe | 97.5% Color 74.7% COD | pH = 12 Electrolysis time = 20 min O ₃ flow rate = 1 g/hr Current density = 9.6 mA/cm ² | Real cardboard factory wastewater |

peroxide may be formed. Thus, according to Eqs. 46–49¹⁰⁷ both OH[•] and SO₄^{•-} are generated.



Enhanced electro-Fenton processes have been created to advance the treatment process regarding water reclamation. Peroxi-coagulation, sonoelectro-Fenton, photoelectron-Fenton, solar photo-electro-Fenton, bioelectro-Fenton, and Fered-Fenton are only a few examples^{108,109}. The efficacy of these EF-based techniques as a robust wastewater treatment approach has absorbed the interest of academic researchers and industry companies, who are constantly investing in developing these technologies. A number of challenges must be overcome in order to make this process more attractive for large-scale industrial use, such as the need for a suitable electrolyte, high energy usage, very low pH, and the possible evolution of hazardous intermediates in the treated water, among other things¹¹⁰.

The EF method is believed to be environmentally friendly despite its drawbacks, as it does not generate any harmful byproducts such as secondary pollutants¹¹¹. It is possible, however, that harmful organic or inorganic compounds will be formed if the overall pollutant mineralization goal is not met. However, it should be noted that the focus of this section is on the combination of EC with H₂O₂, Fenton, and/or electro-Fenton processes. Thus, the application of solo electro-Fenton processes, in which only mixed metal oxides (MMO), graphite, graphene, boron doped diamond (BDD), and dimensionally stable anodes (DSA) electrodes are used (which are very similar to the electrooxidation processes) are not mentioned here. Nevertheless, electro-Fenton process using mentioned materials as electrodes are proven to be a great candidate for removing difficult-to-degrade pollutants and also contaminants of emerging concerns such as pharmaceuticals¹¹² like Metoprolol¹¹³, Levofloxacin¹¹⁴,

Ciprofloxacin¹¹⁵, Tramadol¹¹⁶, Chloroquine¹¹⁷, Imatinib¹¹⁸, Sulfathiazole¹¹⁹, 5-fluorouracil¹²⁰, Acetaminophen¹²¹ and pesticides^{122–125}.

For the treatment of distillery wastewater conducted by Asaithambi et al.¹²⁶, a variety of parameters, including current density, pH, and concentration of H₂O₂ were measured. Dubey et al.¹²⁷ conducted an experimental work using H₂O₂ assisted EC process by both Al and Fe electrodes for the treatment of the effluent from a biodigester. They concluded that iron electrodes were a better choice and the cost estimation for the operation was 1.56 US \$/m³ in this case. Electro-Fenton was also used for treating an actual brewery effluent with high organic content and toxicity and great results were achieved (90% TOC removal)¹²⁸.

Because the main focus of this paper is whether hybrid EC processes are useful for removing emerging contaminants, it should be mentioned that the peroxi-EC process was found to be useful for removing Azithromycin from synthetic wastewater¹²⁹. In terms of disinfection of bacterial contamination, peroxi-EC was more successful than the mere EC process¹³⁰, which was also shown for cyanobacterial removal¹³¹. By using a sacrificial iron anode, air breathing cathode, and Ti/IrO₂ electrode, An et al. demonstrated that the simultaneous EC-EF system produced iron ions and H₂O₂ in situ, thereby eliminating the need for chemical reactants to be purchased, transported, and stored (Fig. 10a). Various current intensities were applied to alter the release rate and quantity of oxidizing reagents to suit different requirements for degrading persistent organic contaminants. As it can be seen in Fig. 10b, increasing current intensity from 7 mA to 35 mA, favored microcystins' removal up to 99%. A large number of coagulants nearly completely covered all algae cells in their SEM images (Fig. 10c). Various Fe valences resulted in different floccule morphologies. In addition, air bubbles produce shear forces that scatter floccules and shape them rounder and more regularly (Fig. 10c). In aerated ECs, coagulation performance was better due to three reasons: 1) Aeration improved contact rates between the cyanobacterial cells and the coagulant, resulting in better removal efficiency; 2) Unaerated ECs had slower pH increases that weakened charge neutralization and lowered cyanobacterial cell removal; 3) Nanosized Fe(III) floccules with higher specific surfaces formed in-situ and easily adsorbed to algal surface.

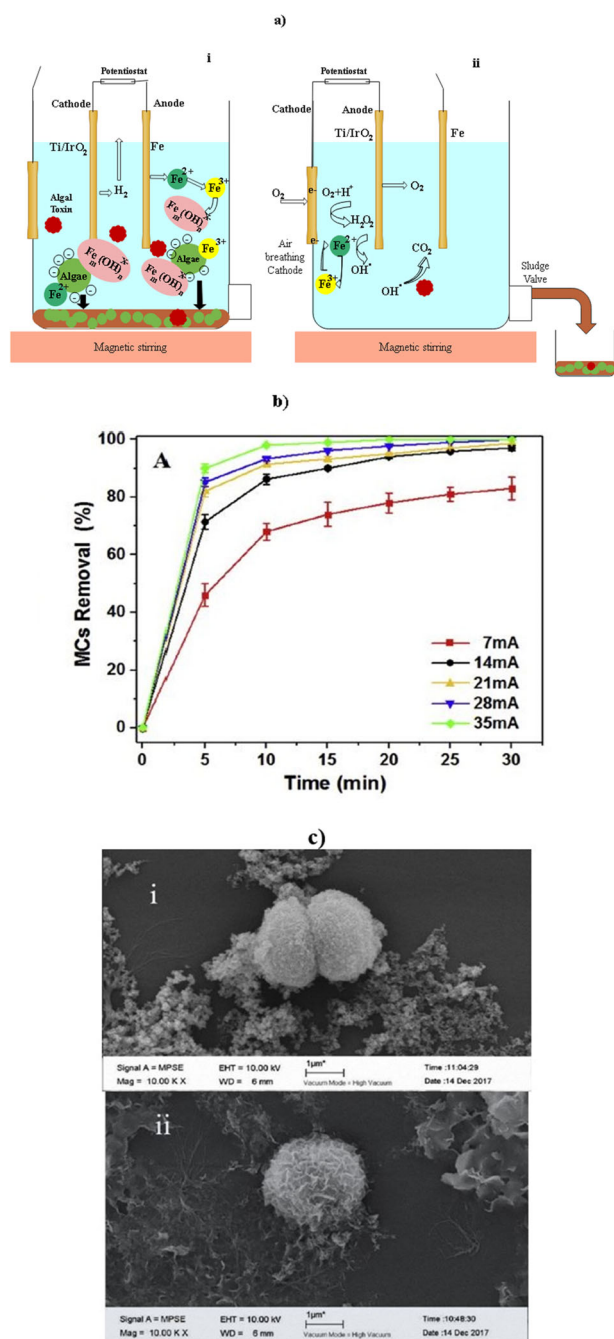


Fig. 10 EC-Fenton mechanism in EC-Fenton reactor and microcystins degradation. **a** EC-Fenton mechanism in (i) EC approach and (ii) EF approach. **b** The degradation of microcystins within 30 min at various current intensities (Conditions: pH = 3, 0.05 M Na₂SO₄, stirring rate: 600 rpm). **c** SEM images of the generated algal flocs at distinct magnifications: Flocules produced under aerated condition (i); Flocules produced under unaerated state (ii). Reproduced with permission from ref. ¹³¹. Copyright © 2019 Elsevier Ltd.

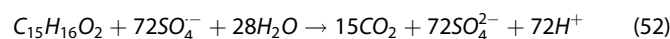
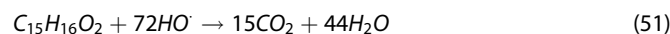
The removal of Perfluorocarboxylic acids (PFCAs) and Perfluorooctanoic acid (PFOAs), which are commonly consumed in domestic products, semiconductor industrial, as a surfactant in medical activities, fire retardant, and metal coating, was conducted using a combination of aerated EC and modified peroxi-coagulation¹³². It was shown that aeration significantly increased the EC process's performance. Because hydroxyl radicals are created on site, peroxi-coagulation excels aerated EC. Figure 11

illustrates their suggested pathway of mixed valent Fe-oxide formation and Fe-hydroxide phases at different pH values during EC process. However, pourbaix diagram of iron confirms that pH conditions change during treatment.

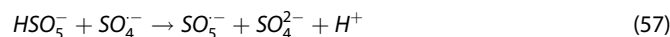
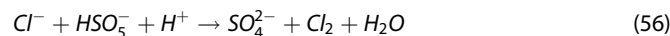
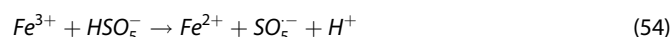
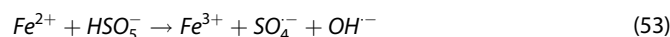
The removal rate of atorvastatin, a drug for lowering the cholesterol, by the peroxi-coagulation process with Fe electrodes was higher than the solo EC process¹³³ and so was for cefixime, an antibiotic, with both iron¹³⁴ and aluminum electrodes¹³⁵, and also for the degradation of lamivudine, which is an antiretroviral drug¹³⁶. The amount of electrode consumption also can be calculated according to Eq. 50¹³³:

$$C_{\text{electrode}} = \frac{it_{\text{process}}M_W}{zFV} \quad (50)$$

Bisphenol A as another hazardous compound and endocrine-disruptor was successfully removed by peroxi-coagulation process^{137,138}. Akbari et al.¹³⁷ proposed Eq. 51 and Eq. 52 for the mineralization of Bisphenol A by hydroxyl radical and persulfate, respectively. They also suggested recycling electrochemical sludge in its suspended form as a promising way to decrease the generated sludge since it was seen that hydrogen peroxide and persulfate were effectively activated by the sludge.



In another study¹³⁹, the degradation of pentachlorophenol applying electrochemical activation of peroxodisulfate (PDS), H₂O₂ and peroxomonosulfate (PMS) with iron electrodes demonstrated that PMS was more effective than two other oxidants (Eqs. 53–57); because chloride ions can also activate PMS to produce oxidizing agents including hypochlorite (HOCl) and chlorine gas.



Persistent mixed industrial wastewater from numerous industries such as cotton, textile, rubber, chemical plants, oil, and plastic factories was treated by EC-EF system¹⁴⁰. It was found that heterogeneous EF catalysts made from alkali modified laterite soil were superior to those made from pure laterite regarding removal performance. COD removal rate after 1 h of the EF treatment was 54.57%, while it increased to 85.27% after 2 h of EC-EF treatment. Conducting EC after EF process was a better option in spite of similar performance. The reason was mentioned to be because of the need to nullify neutralization after EF process not only mineralization efficacy was high, but improved biodegradability, and lower sludge formation as well. Also, it should be noted that the higher current density would not favor the removal rate necessarily as the concentration of hydroxyl radical can be decreased because of side reactions¹⁴¹.

Also, the combination of EF/Fenton and EC process was proven to be successful for treating different types of wastewaters. Gunawan et al.¹⁴² treated a yarn dyed wastewater (also done by¹⁴³) in a batch mode using EC to lessen Total Suspended Solid (TSS), followed by Fenton process in a sequential continuous system to reduce COD level. In another study, different electrochemical oxidation processes were examined for the treatment of an actual textile effluent and the sequential process of EC-EF was chosen as the best method, regarding higher degradation effectiveness and lesser consuming energy¹⁴⁴. Kumar et al.⁹³

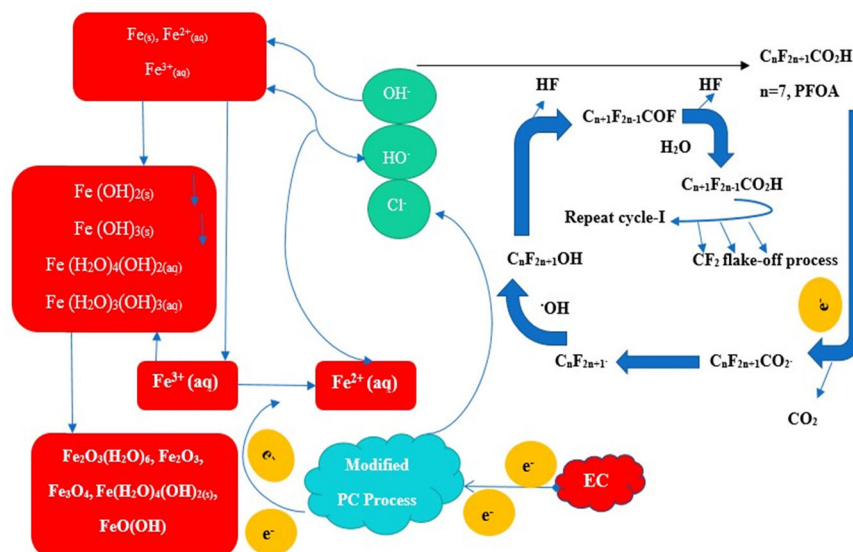


Fig. 11 A possible mechanism for PFOAs removal by EC process. The schematic shows mixed valent Fe-oxide formation and Fe-hydroxide phases at different pH using EC process of PFPAs degradation. Reproduced with permission from ref. ¹³². Copyright © 2021 Elsevier Ltd.

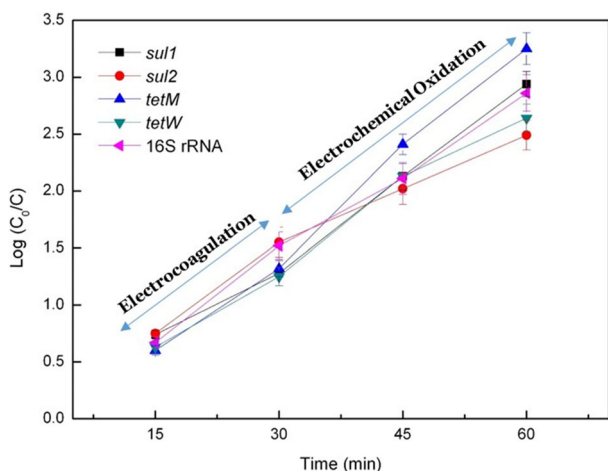


Fig. 12 ARGs and 16S rRNA reduction efficiency from swine wastewater by combined EC/EF processes. The removal efficiency of sequencing EC/EF processes for selected ARGs were 2.94 logs (*sul1*), 2.49 logs (*sul2*), 3.25 logs (*tetM*), 2.64 logs (*tetW*), and 2.86 logs (16S rRNA) during 60 min. Reproduced with permission from ref. ¹⁴⁷. Copyright © 2021 Mary Ann Liebert, Inc.

treated composite wastewater using aerated EC and modified peroxi-coagulation processes. The improved peroxi-coagulation process has a much greater efficacy than aerated EC due to the superior oxidant activity of in-situ produced hydroxyl radicals. For treating landfill leachate, simultaneous EC and EF achieved the concurrent removal of organics and ammonia¹⁴⁵. Electrodisinfection of primary and secondary effluents from municipal wastewater treatment plants showed that EC with iron electrodes was better for the treatment of the primary effluent, with total removal of coliphages, eukaryotes and *E. coli.*, while EF with BDD anode was better for the treatment of the secondary effluent¹⁴⁶. Conducting EF at pH of 7 had almost same results as that of EF at pH 3. Chen et al.¹⁴⁷ applied integrated EC/EF processes to degrade selected antibiotic resistance genes namely *sul1*, *sul2*, *tetM*, *tetW* and 16S rRNA from Swine Wastewater. As can be seen from Fig. 12, they found that removal efficiency of selected ARGs accounted for 2.94 logs (*sul1*), 2.49 logs (*sul2*), 3.25 logs (*tetM*), 2.64 logs (*tetW*), and 2.86 logs (16S rRNA) during 60 min which is shown

sequencing EC/EF processes were more efficient than standalone EC. The results proved that EC as a pretreatment could remove bacterial cells through adsorption and enmeshment of precipitated flocs to enhance the removal efficiency in integrated processes. Sequential EC/EF process was proposed as the best disinfection treatment. The details of reviewed papers are mentioned in Table 3.

Because wastewater contains inorganic ions, the Fenton and photo Fenton processes become more complicated. With the existence of anions such as NO_3^- , PO_4^{3-} , SO_4^{2-} , and Cl^- the reaction rate of H_2O_2 with ferrous ion is different¹⁴⁸. To decrease the production of reactive hydroxyl radicals and the removal rate of pollutants, these ions can either form indirect chloride ion-scavenging HO^\cdot or less reactive complexes with iron and ferric ions¹⁴⁹. Chloride ions, chlorate and perchlorate oxyanions, (oxy) chlorinated radicles, chloramines, hydroxyl amines, and trihalo-methanes can all be produced as well as refractory organo-chlorinated byproducts as undesirable byproducts¹⁵⁰.

HYBRID ELECTROCOAGULATION-UV

Reactive oxygen species are generated when a semiconductor surface is exposed to high-intensity light, which is used in photocatalysis¹⁵¹. When a semiconductor is stimulated using light of energy larger than or equivalent to its band gap, production of electron-hole pair occurs. As a result, reactive oxygen species can be produced in the aqueous phase when oxygen is present¹⁵² (Fig. 13). An interaction occurs between UV light and the contaminant molecule, thus the molecule is degraded and mineralized as a direct result. Hydrogen peroxide that can boost the release of hydroxyl radicals, enhances this direct photolysis even more^{153,154}. Photoelectrochemical systems consist of an electrolytic reactor containing an illumination source for the photoanode, a photocatalyst and an electrical power supply or potentiostat. Even though, using various reactors including stirred tank and flow reactors, as well as different light sources within/outside the cell, such as a quartz window, UVC (300 nm > λ_{max}), and UV/Vis lamps (λ_{max} from 320 to 400 nm) have all been considered. Xe lamp can replicate sunlight to allow UV/solar irradiation on the anode surface. In photo-Fenton, Fenton reagents are irradiated with UV-Visible light. In this process hydroxyl radical generation is enhanced within two mechanisms: (i) the reproduction of active Fe^{2+} ions by reducing Fe^{3+}

Table 3. Peroxi-coagulation, hybrid EC/Fenton, or EC/EF processes for treating water and wastewater.

| Ref. | Wastewater/Pollutant | Type of hybrid process and Electrode materials in EC | Removal rate | Comments |
|------|--|--|--|--|
| 209 | Real pasta and cookie processing wastewater | Peroxi-coagulation (Al) | 90% COD 96% BOD 99.9% Fecal coliforms | pH = 4 Current density = 18.2 mA/m ² H ₂ O ₂ = 1.5 ml/L |
| 147 | Antibiotic Resistance Genes | EC-EF | 2.49–3.25 logs for intracellular ARGs 3.23–4.38 logs for extracellular ARGs | EC time = 30 min EF time = 30 min Neutral pH Current density = 20.0 mA/cm ² |
| 139 | Pentachlorophenol (PCP) | EC(Fe)-Peroxomonosulfate (PMS) | 75% PCP | pH = 4.5 Current intensity = 90 mA EC time = 30 min PMS = 9.9 × 10 ⁻⁵ M Removal performance order: EC/PMS > EC/PDS > Peroxi- EC > EC |
| 210 | Wastewater containing Malathion | EC(Al)-H ₂ O ₂ | 96% Malathion | Voltage = 20 V Time = 200 min |
| 211 | Aqueous solution containing Metronidazole | Peroxi-coagulation (Fe) | 100% Metronidazole | pH = 7 Time = 40 min Metronidazole concentration = 50 mg/L Voltage = 25 V H ₂ O ₂ = 0.01 Mol/l |
| 137 | Bisphenol A (BPA) | Peroxi-coagulation (Fe) EC(Fe)-Persulfate | 84% BPA 72% BPA | pH = 3 Current density = 0.8 mA/cm ² H ₂ O ₂ = 8 mM pH = 5 PS = 6 mM Current density = 1 mA/cm ² |
| 212 | Distillery effluent | Peroxi-coagulation and peroxi-photo-coagulation (Fe) | 95.85% COD 100% Color | Current density = 0.3 A/dm ² [H ₂ O ₂] = 250 mg/dm ³ O ₃ -EC performed better than peroxi-coagulation |
| 133 | Atorvastatin | Peroxi-coagulation (Fe) | 82% Atorvastatin 77% TOC | pH = 3 Electrolysis time = 30 min H ₂ O ₂ = 250 mg/L Current density of EC = 2.5 mA/cm ² Energy consumption = 1.373 kWh/m ³ Overall cost = 0.159 €/m ³ |
| 213 | Aqueous perfluorooctanoic acid (PFOA) | Peroxi-coagulation (Fe) | 99% PFOA | pH = 3.77 Electrolysis time = 40 min H ₂ O ₂ = 0.66 g/L Current density of EC = 37.5 mA/cm ² Operational cost = 2.8\$/kg PFOA removed |
| 146 | Primary and secondary effluents from municipal wastewater treatment plants | EC(Fe)-Electro-Fenton (BDD) | Inactivation of all the active microbiota | Natural pH (EC) EC time = 30 min Current density of EC = 200 A/m ² pH (EF) = 3 EF time = 30 min |
| 214 | Wet-spun acrylic fibers manufacturing effluents | Peroxi-coagulation (Fe) | 80.9% EC ₅₀ | pH = 5 Electrolysis time = 30 min H ₂ O ₂ = 10 mM Current density of EC = 35.7 mA/cm ² |
| 141 | Cu-EDTA synthetic solution | Peroxi-coagulation (Fe) | 98.2% Cu-EDTA | pH = 3 Electrolysis time = 30 min Current density = 72.92 A/m ² H ₂ O ₂ = 49.4 mM |
| 131 | Cyanobacteria and cyanotoxins | EC(Fe)-EF(Ti/IrO ₂) | 100% Cyanobacterial 100% Microcystins 100% TOC | pH = 8 EC time = 40 min EF time = 30 min |

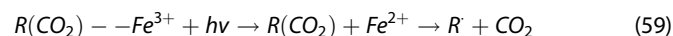
Table 3 continued

| Ref. | Wastewater/Pollutant | Type of hybrid process and Electrode materials in EC | Removal rate | Comments |
|------|--------------------------------------|--|---|---|
| 215 | Palm oil mill effluent | Peroxi-coagulation (Fe) | 71.3% COD 96.8% Color 100% TSS | Current intensity of EC = 100 mA Current intensity of EF = 35 mA pH = 4.4 Current density = 40.21 mA/cm ² H ₂ O ₂ = 0.5 g/L Electrolysis time = 45.67 min |
| 134 | Crude drug effluent | Peroxi-coagulation (Fe) | 99.98% cefixime | pH = 4.53 Current density = 136.5 A/m ² H ₂ O ₂ = 12 ml/L Time = 60 min Fe performed better than Al |
| 216 | Ciprofloxacin (CIP) | EC(Al)-Persulfate | 90% CIP | pH = 7 Electrolysis time = 40 min Current density = 2.75 mA/cm ² PS = 0.84 mM |
| 217 | Canola oil refinery wastewater | Peroxi-coagulation (Fe) | 77% CODs 86% DOC | Current density = 13.66 mA/cm ² H ₂ O ₂ = 3 mL (30%)/250 mL wastewater Electrolysis time = 7 h |
| 144 | Real textile wastewater | EC(Fe)-Peroxi-coagulation/EF (BDD) | 97% TOC 100% Turbidity 100% Color | pH = 3 Current density of EC = 10 mA/cm ² Energy consumption of EC-EF = 0.45 kWh/kgTOC Energy consumption of Peroxi-coagulation = 1.9 kWh/kgTOC |
| 135 | Cefixime | Peroxi-coagulation (Al) | 92.25% Cefixime | pH = 5.26 Electrolysis time = 60 min H ₂ O ₂ = 0.63 mg/L Current intensity = 0.3 A |
| 138 | Bisphenol A (BPA) | Peroxi-coagulation (Fe) | 80.48% BPA | pH = 2 Electrolysis time = 45 min H ₂ O ₂ = 125 mg/L Current density = 0.11 mA/cm ² |
| 145 | Leachate concentrate | EC(Fe)-EF(DSA) | 68% TOC 57% COD 60% Ammonia | pH = 7 Current density = 30 mA/cm ² Energy consumption = 67.8 kWh/kgCOD Novel dual-anode system Treatment effectiveness: EC-EF > EF > EC |
| 127 | Rice gain based biodigester effluent | Peroxi-coagulation (Fe/Al) | 98.3% COD 83.6% Color | pH = 7.3 H ₂ O ₂ = 120 mg/dm ³ Current density of EC = 114 A/m ² Energy consumption = 1.5 Wh/dm ³ |
| 105 | 2,4,6-trinitrotoluene (TNT) | EC(Fe)-Persulfate | 88% TNT | pH = 5 PS = 0.3 mg/L Current density = 1.75 mA/cm ² Electrolysis time = 45 min |
| 132 | Perfluorocarboxylic acid | Modified peroxi-coagulation (Fe) | 90% PFCA 87% TOC 71% defluorination | pH = 3.6 Current density = 78 mA/m ² Electrolysis time = 60 min H ₂ O ₂ = 50 mM |

(UV < 580 nm), and (ii) the photolysis of hydrogen peroxide (UV < 310 nm)¹⁵⁵.

When Peroxi-EC process is integrated with UV photolysis, the number of photoactive sites has a positive impact on the EC process. Fe(OH)²⁺ photoreduction and the photodecomposition

of complexes produced during Fe³⁺ reactions help increasing the rate of synthesis of OH[•] (Eqs. 58–59):



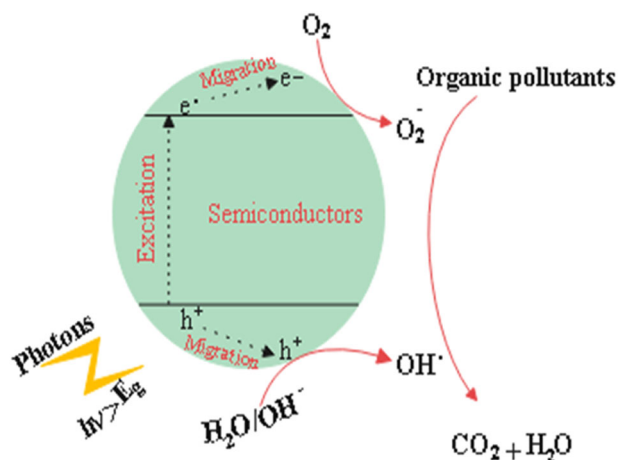


Fig. 13 Schematic diagram of a semiconductor's surface during photocatalytic oxidation. In this mechanism, when a semiconductor is irradiated by light, electron-hole pairs are generated. An electron (e^-) will jump into the conduction band, leaving a hole (h^+) in the valence band. Then, the photo-generated electron-hole (e^-h^+) pairs are separated, and migrate to the surface or interface of the photocatalyst. Reprinted with permission from ref. ²²⁶. Copyright © 2015 Elsevier Ltd.

The sequential EC and photooxidation methods were used to remediate high-strength tannery effluents^{156–158}. Aside from direct photolysis of organic contaminants, UV had a significant impact on degradation pathways. Coagulation formed flocs that helped remove total chromium from the photooxidation process, and the simultaneous oxidation of organic molecules and reduction of Cr (VI) occurred. Iron electrodes were found to be improper due to production of Iron(II) sulfide flocs resulted in turbid appearance of the treated wastewater. UV irradiation and EC with aluminum electrodes can also remove microbial content and turbidity from urban treated wastewaters at low current densities¹⁵⁹. When the pH during the process stayed around 8, insoluble aluminum hydroxides formed. Also, free and mixed chlorine disinfectants were found in wastewater. Urban wastewater contains chlorides that could be oxidized on the anode surface, generating hypochlorite. Electrogenerated hypochlorite reacts with wastewater ammonium to make chloramines. Both species had disinfection abilities and eliminated *E. coli*. The interaction of UV light with *E. coli*'s cell membrane enhanced the eradication of bacteria. UV light during EC promoted free radical production from oxidizing species and they boosted *E. coli* removal performance and also favored dissolution of the sacrificial electrode, enhancing turbidity removal.

The simultaneous EC-photo-Fenton was performed in order to treat steel industry effluent using a single reactor with a UV-C lamp and aluminum electrodes¹⁶⁰. Yahiaoui et al.¹⁶¹ observed a decrease in the removal of metribuzin when H_2O_2 was introduced to a UV aided EC system. The authors believe that increasing turbidity might be the cause. Módenes et al.¹⁶², on the other hand, treated tannery wastewater through a batch mode incorporation of solar photo Fenton assisted EC and reached significant enhancement in the reduction of COD and also sludge generation. In a work conducted by Farhadi et al.¹⁶³ EC, photoelectrocoagulation, peroxi-coagulation and peroxi-photoelectrocoagulation processes were compared in terms of treating a pharmaceutical wastewater.

The integrated EC-Fenton/photo-Fenton processes have been shown to be a promising treatment to nearly complete removal of the phenolic compounds in oil refinery wastewater¹⁶⁴. Also, adding UVA-LED photo-Fenton as post-treatment after the EC process for the treatment of mature landfill leachate was very

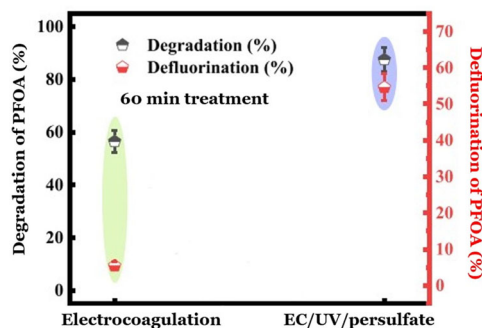


Fig. 14 The degradation and defluorination of PFOA in EC and combined EC/UV/persulfate (PFOA concentration = 100 mg/L, current density = 12.5 mA/cm², pH = 6). Under conditions of PFOA concentration = 100 mg/L, current density = 12.5 mA/cm², and pH = 6. PFOA removed only 56.4% by EC, while it soared to 87.5% using EC/UV/persulfate during 60 min. In addition, defluorination of PFOA for such combined process was 60% compared to 6% EC.

successful¹⁶⁵. Zhang et al.¹⁶⁶ investigated the efficacy of EC standalone and persulfate-assisted and UV-activated electrocoagulation technique using Fe foam electrode for treating PFOA from water. As illustrated in Fig. 14, they observed that EC only reduced 56.4% of PFOA, whilst it soared to 87.5% during 60 min by EC/UV/persulfate. On the other hand, defluorination of PFOA for such combined process (60%) accounted for 10 times higher than EC (6%). This because of sulfate radical generation by the activation of ferrous ions under the synergy of UV light and electric energy. The summary of reviewed articles is given in Table 4.

Synergistic effect (SE) (also called enhancement factor¹⁶⁷) plays a crucial role in developing a hybrid approach for wastewater treatment¹⁶⁸. Synergy is the amplified result produced by the combination of two or more processes or variables, as opposed to the sum of their individual impacts. Equation 60¹⁶⁹ can be employed to get the SE by comparing the integrated process's pollutant removal efficiency to the sum of the separate processes' pollutant removal rate.

$$SE = \left(\frac{k_{AOP+EC}}{k_{AOP} + k_{EC}} - 1 \right) * 100 \quad (60)$$

Where k_{AOP+EC} , k_{AOP} , and k_{EC} are the rate constants of the hybrid process, the individual AOP, and the individual EC process, respectively. When the SE value is positive, it indicates a positive synergistic effect. The combined process is more effective because of: i) enhanced mass transfer and electrode activation, ii) no passive layer on the electrode surface, and iii) release of more hydroxyl radicals.

Another important issue is the scale-up of the hybrid processes, as it is a vital aspect in the practical use of every process for drinking/waste water treatment¹⁷⁰. The surface area to volume ratio (S/V) is a critical scale-up factor. It is important to note that the electrode area has a significant effect on the current density, the rate of cation dosing, bubble development, and the bubble paths' length. According to Mameri et al.¹⁷¹, the ideal current density declines with increasing S/V. However, there was little information available on the S/V ratio. Following dimensionless scale-up criteria have been recognized for EC to progress from experimental to industrial levels¹⁷²:

- Reynolds number for describing the fluid flow regime
- Weber number for indicating the surface tension
- Froude number as an index of buoyancy
- Geometric resemblance
- Gas saturation comparison

Table 4. Hybrid EC-UV for CECs removal from water/wastewater.

| Ref. | Wastewater/Pollutant | Process/Electrode materials | Removal rate at optimal conditions | Comments |
|------|--|---|---|---|
| 161 | Ground water | Anode: Fe Cathode: SS | 95% Metribuzin | pH = 6 Current density = 18 A/m ² |
| 166 | Simulated wastewater | EC (Fe)/ persulfate/ UV | 87.5% PFOA | Initial pH at photolysis = 3 Initial pH at EC = 8.3 Fe ²⁺ = 0.4 g/L H ₂ O ₂ = 15 g/L |
| 218 | Pulp and paper wastewater | SS | 85% COD | pH = 7 Current density = 50 mA/cm ² |
| 159 | Urban wastewater | Anode: Al Cathode: SS | Complete removal of E.Coli | pH = 8 Current density = 17.3 A/m ² |
| 219 | Tartrazine (dye pollutant) | EC(Fe) + photoelectro-Fenton (Ti/IrO ₂ -RuO ₂) | 60% COD 100% color | pH = 6.3 EC time = 12.5 min Photoelectro-Fenton time = 360 min Current intensity = 200 mA |
| 220 | Hydroquinone | Anode: Al Cathode: SS | 94% TOC | pH = 4 Current density = 15 mA/cm ² |
| 221 | Lithographic wastewater | Fe | 74.43% TOC | EC time = 20 min Photocatalysis time = 45 min |
| 164 | Oil refinery wastewater | Fe | 88% TOC | EC time = 20 min Photolysis time = 60 min Current density = 40 mA/cm ² Fe ²⁺ /H ₂ O ₂ = 1:66 H ₂ O ₂ = 306 mg/L |
| 156 | Tannery wastewater | Fe | 80% COD 95% color 98% Turbidity | pH = 4 photoelectrolysis time = 120 min H ₂ O ₂ = 6 g/L Current density = 25 mA/cm ² |
| 222 | Olive oil mill wastewater | Fe/Al/SS | 91.7% TOC | pH = 7 electrolysis time = 600 min |
| 160 | Steel industry effluent | Al | 100% Phenol 97% COD | Photoelectrolysis time = 25 min pH = 4 Current density = 1.5 mA/cm ² Energy consumption = 29.15 kWh/kgCOD EC/photo Fenton hybrid process |
| 223 | Micropollutants in reverse osmosis brines | Al | 63% DOC | Photoelectrolysis time = 240 min pH = 5.5 Current density = 9.4 mA/cm ² |
| 134 | Cefixime from crude drug effluent (after treating by peroxi-coagulation) | Fe | 33% TOC (30% with sunlight) | EC time = 60 min Current density of EC = 136.5 A/m ² Photocatalysis time = 12 hr TiO ₂ = 0.25 g/L |
| 158 | Tannery wastewater | Al | 99.52% COD 100% Cr 98.27% Sulfide | pH = 4 Photoelectrolysis time = 20 min Current density = 1.5 mA/cm ² |
| 224 | Pharmaceutical wastewater | Fe | 41% TOC | pH at EC = 7 pH at photocatalysis = 10 EC time = 120 min Photocatalysis time = 240 min ZnO = 1 g/L |
| 157 | Tannery wastewater | Al | 94.1% COD | pH = 10.78 EC time = 5 hr UV time = 3 hr Current density = 12 A/cm ² Energy consumption = 314.28 kW h/m ³ |
| 136 | Lamivudine | Photoperoxi-coagulation (SS) | 96% COD | pH = 3 H ₂ O ₂ = 150 mg/L Current intensity = 4.6–4.8 mA/cm ² EC time = 120 min |
| 225 | Distillery industrial wastewater | Fe | 100% Color 95.6% COD | pH = 7 reaction time = 4 hr Current density = 0.175 A/dm ² Energy consumption = 6.97 kW h/m ³ EC + sono+UV Synergy index (Eq. 60) = 20% |

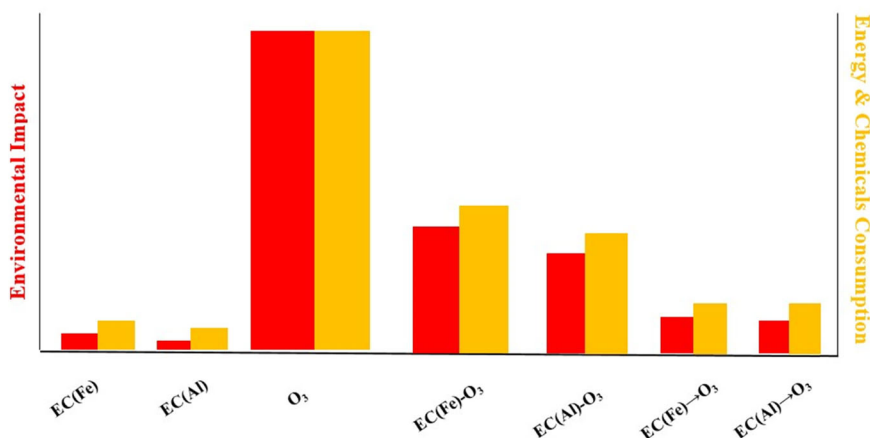


Fig. 15 Comparison of EC, O₃ and their combination through LCA. Reproduced with permission from ref. ¹⁹⁰. Copyright © 2021 Elsevier Ltd.

Nevertheless, scaling-up of EC-integrated processes from laboratory to industrial scale is of course more complicated and needs more research.

LIFE CYCLE ASSESSMENT OF INTEGRATED EC/AOPS PROCESSES: THE WAY FORWARD

Urban water infrastructure, including wastewater treatment plants (WWTP), has extensively been quantified using life cycle assessment (LCA) during the last 20 years¹⁷³. This shift from a pollution-removal paradigm into one that incorporates resource recovery is where LCA can play a significant role in evaluating the environmental sustainability of new technologies and processes and capturing trade-offs across various environmental categories¹⁷⁴. Life cycle assessment is an effective decision support tool for water sector strategic planning because of its quantitative nature as an environmental evaluation approach¹⁷⁵. Due to the involvement of multiple processes that are interdependent (e.g., effluent from one process is often influent for the next), elucidating the environmental implications of WWTP variations via LCA can be rather complicated. Furthermore, as the paradigm change continues, WWTPs are becoming multi-functional, with aims that go beyond standard effluent quality criteria (BOD₅, TSS, etc.) to include resource recovery, energy management, emerging pollutant removal, and so on. Addressing such complications in the application of LCA to wastewater management can be aided by the creation of a set of best practices customized particularly to this field of research¹⁷⁶.

Ozonation is one of the most studied technologies for the removal of contaminants of emerging concern. It is thought to be more environmentally friendly than UV in terms of pharmaceuticals' removal¹⁷⁷. According to some research, it has a greater environmental impact than it prevents. Hoibye et al.¹⁷⁸, on the other hand, determined that the prevented effects are over-estimated since these technologies remove more than simply the identified pollutants. Thus, when more compounds are included, ozonation produces less consequences than it avoids, a finding validated by another study¹⁷⁹. In contrast to Tarpani and Azapagic¹⁸⁰, Rahman et al.¹⁸¹ concluded that ozonation had a lower impact on global warming and ozone depletion than activated carbon. Because chemical and energy production contribute more to these categories, the disparity may be due to differing energy mixes. According to Li et al.¹⁸², ozonation had one of the lowest impacts on the categories of acidification potential, human toxicity non-cancer, global warming potential, fossil depletion, smog, and eutrophication. Rodriguez et al.¹⁸³ suggest that lower O₃ intakes are preferable if a balance between toxicity reduction and greenhouse gas emissions is desired.

When employing light-based processes, the source of photon production determines total environmental implications. Both natural and simulated solar light have limited efficiency, and as a result, the treated effluent discharge has significant consequences. In a comparison of UV-A and UV-C without the use of H₂O₂ or any catalyst, Foteinis et al.¹⁸⁴ determined that UV-C is about three times less dangerous than UV-A. That is because it is substantially more efficient for eliminating micropollutants, while requiring more power. When H₂O₂ is added to the UV-C treatment, the environmental footprint is decreased by up to 88%. In their study, UV light (both UV-A and UV-C) had the greatest influence on the endpoint category of human health, followed by resources, and finally ecosystems. TiO₂ was also used to evaluate the possibilities of improving the UV-A process. While it was discovered that UV-A was the least environmentally friendly option when compared to UV-C, and that UVC + H₂O₂ was even more environmentally friendly, when UVA is combined with TiO₂, the environmental performance of the process outpaces that of UV-C with or without H₂O₂ (despite TiO₂ having a slightly larger footprint than H₂O₂).

LCA has not been performed on Fenton reaction without light for micropollutant and emerging contaminants' elimination. Most LCA research were conducted on photo-Fenton and so energy usage is once again an important issue. Solar photo-Fenton is more sustainable than photo-Fenton using artificial/simulated light since it does not require energy (note that external electricity may be needed for equipment, such as pumps). Electro-Fenton has higher repercussions than solar photo-Fenton and solar photo-electro Fenton. After energy generation, H₂O₂ production is the greatest strain in photo-Fenton process. Ioannou-Ttofa et al.¹⁸⁵ found that NaOH contributed most to total effects, followed by H₂O₂ and H₂SO₄. Solar photo-Fenton at stoichiometric H₂O₂ is the most environmentally sound solution, with the smallest footprint. Another study¹⁸⁶ found that integrating solar photo-Fenton with nanofiltration boosts operating energy consumption while reducing chemical use, lowering total life cycle impacts. Solar photo-Fenton results in reduced loads since it removes micropollutants faster, however at larger scale the findings may be influenced by the decreased reaction volume. Farre et al.¹⁸⁷ found that the best option in environmental terms was the coupling of solar photo-Fenton with the biological treatment, which resulted in less or up to 50% less impacts than photo-Fenton in all categories except for aquatic eutrophication potential. Only one publication¹⁸⁸ on the life cycle assessment of the electrooxidation process was found, in which it was claimed that electrooxidation is an environmentally viable method since it generates reduced total environmental consequences, including CO₂ emissions to the atmosphere.

In terms of electrocoagulation process, there is very few studies about LCA, one of which focused on life cycle assessment for the

selection of electrodes for the treatment of a paper industry wastewater¹⁸⁹. Factors like environmental impact of the electrode production, the consumption of materials and the power usage were considered and Fe-Fe arrangement was found to be more environmentally friendly. Moreover, only one paper was found about LCA for the combination of electrocoagulation and advanced oxidation processes (Fig. 15). Ahangarnokolaei et al.¹⁹⁰ studied life cycle assessment of sequential and simultaneous combination of electrocoagulation and ozonation for the treatment of a textile wastewater. ReCiPe midpoint and endpoint approaches were used to compare lab-scale processes. As can be seen in Fig. 15, due to decreased energy usage, the EC process with Al electrodes had the lowest environmental effect. In terms of human health, O₃ had a stronger effect than EC with Al electrodes. High ozonation time makes simultaneous combination (EC-O₃) environmentally harmful, but in sequential combination, O₃ treatment duration was very short and environmental consequences dropped dramatically. LCA demonstrated that the sequential combination of EC and O₃ is the most sustainable textile wastewater treatment process and can be implemented industrially.

OUTLOOK AND SUMMARY

The combination of AOPs with EC is helpful for treating water and wastewater and markedly improves performance. It is both extremely promising and very challenging. This review focused on this topic, giving the researchers a roadmap for covering the knowledge gap and tackling the obstacles, and investigating different aspects. Various mechanisms, pathways for pollutant removal, kinetic models, properties of experiments, and also scale-up issues, have been reviewed in this paper. These features differentiate this review from the previous ones as it can be a guideline, especially for those who are vaguely familiar with this topic. More importantly, the chief focus of this review is the application of the hybrid processes for the removal of contaminants of emerging concern, such as pharmaceuticals, pesticides, herbicides, and recalcitrant pollutants. EC-AOP integration has demonstrated to be highly effective for this purpose as a wide range of these contaminants has been removed efficiently by various combinations of EC-AOP. Combining EC with AOPs also led to faster and more cost-effective water and wastewater treatment (less energy consumption). Moreover, the integrated system overwhelms the disadvantages of each method when used standalone, such as lower sludge production.

In order to choose the best course of action, it is essential to determine if existing research on these technologies is tackling the most significant concerns. The following recommendations must be addressed for future studies:

- Future works should be scaled up employing continuous operation and real wastewaters to be presented as an effective and trustworthy water and wastewater treatment method.
- Combination selection could be determined solely on wastewater properties. If the wastewater includes substances that are more resistant and hazardous, especially contaminants of emerging concern, EC followed by an AOP can be a fruitful option.
- Integrated processes can be conducted in a single reactor. External addition of oxidants such as hydrogen peroxide can also increase the initial rate of pollutant degradation. However, it is proposed that heterogenous Fenton process combined with EC is preferable to homogenous Fenton process. Also, aerated EC combined with AOP processes, particularly electro-Fenton, as it has been recognized as an effective method.
- Concerning the combination of EC and electrooxidation, the creation of very stable, inexpensive electrodes with a large

surface area is still an issue. The cost of purchasing and replacing BDD anodes influences the total expenses of the treatment procedure for particularly persistent pollutants.

- CECs' removal potential for reduced absolute toxicity must overcome a shortage of empirical data on its toxicity. Regarding combating micro and nano plastics, there has not been significant research. In terms of pharmaceuticals' removal, peroxi-coagulation as well as the combination of EC with electrooxidation and electro-Fenton has confirmed to be outstanding, and also there has been acceptable results by integrated EC/UV. When it comes to removing pesticides, EC, electro-Fenton, and electrooxidation are proven to be very effective.
- There are few papers that investigated the combination of EC with AOP processes for removing pesticides, especially in terms of ozone and electro-Fenton. Therefore, it is suggested that future works focus on performance of these hybrid processes for removing pesticide-containing effluents.
- More reviews are needed to compare the performance of other EC-integrated processes including adsorption, chemical coagulation, membrane and biological methods for the removal of CECs.
- In the EC processes aided by AOPs, the electrical energy generated by non-renewable resources is demanded. This could result in two main negatives, the high operational costs and the indirect contamination created by the fossil fuels combustion. Using renewable energy sources like wind, solar, and tidal energy or biogas is essential, so authors recommend future works to use these renewable energy sources instead of electricity.
- Regarding life cycle assessment studies, there are still a lot of work to be done as there are only two papers that investigated this issue for hybrid EC/AOPs. According to them, integrating EC with ozone and also solar power are both environmentally friendly and cost effective. Also, ozonation is regarded as one of the most efficient methods for the removal of organic micropollutants in this regard.

Overall, hybrid treatment processes that successfully combine economic feasibility, high removal efficacy, and environmental sustainability are urgently needed in today's world. However, further process optimization, modeling, and scaling-up research is required to verify these integrated processes' reliability for large-scale wastewater treatment.

DATA AVAILABILITY

The authors declare that the data supporting the findings of this study are available within the manuscript.

Received: 18 September 2022; Accepted: 28 February 2023;

Published online: 11 April 2023

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ACKNOWLEDGEMENTS

The support of the Research Vice-Chancellor of Qazvin University of Medical Sciences is wholeheartedly appreciated.

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COMPETING INTERESTS

The authors declare no competing interests.

ADDITIONAL INFORMATION

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